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Proportional Counter Studies
of
Orbital Electron Capture Ratios.

by
P.W. Dougan.

Presented as a Thesis for the degree of Ph.D. in the
Department of Natural Philosophy, Glasgow University,
June, 1961.

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Publications.

The work described in this thesis is to be submitted for publication in the form of three papers:

1. Orbital Electron Capture Ratios in Germanium-71 and Krypton-79.
2. The Decay of Chlorine-36 to Sulphur-36.
3. The L/K-Electron Capture Ratio in Argon-37.

Preface.

The work on orbital electron capture ratios described in this thesis was performed by the author between October, 1958 and December, 1960, in the Department of Natural Philosophy of the University of Glasgow.

The measurements on germanium-71 and krypton-79 were carried out jointly by Dr. R.W.P. Drever and the author. The experimental work and the analysis of the data was shared between both these persons. The special electronic circuits developed as a result of the experience gained in these measurements were designed by Dr. Drever.

The measurements on chlorine-36 and argon-37 were suggested by the author and were carried out by him in conjunction with Mr. K.W.D. Ledingham.

The analysis of the results was divided equally between the author and Mr. Ledingham.

During the course of the measurements on chlorine-36, many useful discussions were held with Dr. Drever.

Summary.

The first chapter of the thesis summarizes briefly the theoretical treatment of orbital electron capture. Particular attention is given to the theoretically-obtained wave function ratios which are available for comparison with experimental values. It is pointed out that, when calculating values of atomic electron wave functions, it is customary not to consider the correlations which must exist among the atomic electrons, and that when such correlations are considered, the theoretical values for L/K-ratios are increased over those obtained from the use of hydrogen-like functions. A comparison of theoretical and experimental values of L/K-ratios is made and it is shown that further experimental work is required in all ranges of atomic number. In particular, the range of atomic number between 15 and 40 is shown to be suitable for investigation.

The second chapter describes the characteristics required in the detection system and outlines the wall-less counter technique used by the author.

The experimental work is described in the next four chapters. Measurements of L/K-ratios in the isotopes germanium-71, krypton-79, chlorine-36 and

argon-37 are described, together with incidental points of counter technique which were clarified in the course of the measurements.

The values of 0.116 ± 0.005 , 0.108 ± 0.005 , 0.112 ± 0.008 and 0.102 ± 0.004 , obtained for the L/K-ratios in these isotopes, are greater than the respective theoretical values of 0.106, 0.101, 0.080 and 0.082 calculated from the ratios of wave functions prepared by Brysk and Rose. The difference between theory and experiment is most marked for isotopes of small atomic number. This is in accord with the predictions of Odiet and Daudel, who evaluated the effect of the electron correlations in light isotopes. Their value of 0.100 for the L/K-ratio in argon-37 is in good agreement with that obtained in the present work.

Capture from the M-shell was detected in germanium-71 and krypton-79, the first direct observation of capture from this shell in isotopes with mass number less than 200. The values of the ratio of M-capture to L-capture in both isotopes was found to be 0.16 ± 0.08 , in reasonable agreement with the ratios of electron density given by Hartree wave functions. Indications that M-capture occurred

also in chlorine-36 and argon-37 were observed, but the evidence cannot be considered to be conclusive.

The final chapter of the thesis summarizes the above results and, after comparison of the available theoretical and experimental values of L/K-ratios, concludes that further theoretical work to evaluate the magnitude of the electron correlations as a function of atomic number is necessary.

There are four appendices. The first contains analytical forms of the wave functions of an electron in the Coulomb field of a point charge, while the second gives some details about the counters used in the measurements. In the third, are some brief remarks about the new electronic units added in the course of the measurements. The fourth discusses measurements undertaken to establish the presence of a positron branch in the decay of chlorine-36. It is shown that the observations are consistent with the existence of a weak branching, of intensity $K/\beta^+ = 1500 \pm \frac{300}{500}$ relative to the K-capture component of the source.

Acknowledgments.

The author would like to thank Professor P.I. Dee for his interest and encouragement throughout the work described in this thesis.

Performance of the work was facilitated by the co-operation of the technical staff under Mr. Lloyd, the workshop staff under Mr. Irvine, the electronic department under Mr. Simpson and the Stores staff under Mr Donaldson.

Several points were clarified in discussion with Dr. G.M. Lewis.

The author would like to express his thanks to both Dr. R.W.P. Drever and Mr. K.W.D. Ledingham, with both of whom his collaboration has been stimulating and productive.

During the period in which the work was carried out, the author was in receipt of a D.S.I.R. Research Studentship.

Chapter I.

Theoretical Aspects of Electron Capture.

The electron capture process is that species of nuclear transformation in which a nucleus of charge Z interacts with an electron, the products of the interaction being the isobar of charge $Z-1$ and a neutrino.

The existence of such a process was first suggested as a consequence of the theory of beta-decay proposed by Fermi (1934), in which the fundamental transformations are

$$(n) \longrightarrow (p) + e^{-} + \bar{\nu} \quad (1)$$

$$(p) \longrightarrow (n) + e^{+} + \nu \quad (2)$$

where n , p , e^{+} , e^{-} , ν and $\bar{\nu}$ denote respectively neutron, proton, positron, electron, neutrino and anti-neutrino and the brackets indicate that the nucleons are bound in the nucleus.

Bothe and Bacher (1936), Yukawa and Sakata (1935) and Möller (1937) pointed out that an alternative form of (2) was

$$(p) + e^{-} \longrightarrow (n) + \nu \quad (3)$$

and that, because of the presence of a reasonable density of bound atomic electrons near the nucleus, such a process should compete with positron emission. Since the latter requires that the energy difference between initial and final nuclear states is at least 1.02 MeV, in nuclei where

the energy available was less than this figure, electron capture as represented by (3) would be the only possible decay mode.

These predictions were verified by the observations of Alvarez (1937, 1938) who detected the K X-rays from a vanadium source which was also a positron emitter. He was able to show that the origin of these X-rays was the atomic re-arrangement subsequent to electron capture from the K-shell.

In formal treatment, such as given by Marshak (1942), Brysk and Rose (1955, 1958) and Bouchez and Depommier (1960), the selection rules and transition probabilities for electron capture are taken over directly from the corresponding expressions for beta-decay, the only changes being that the electron wave functions are those of bound rather than of free particles, the neutrino energy is fixed for capture from a given sub-shell and there is no sum over angular momenta. Thus the selection rules for capture are:

Table 1.

Transition Type	Parity Change	Spin Change
Allowed	No	0,1
First-forbidden	Yes	0,1,2
n-times forbidden ($n \geq 2$)	$(-1)^n$	$n, n+1$

For allowed transitions, theory shows that the ratio R of L- to K-capture is given by

$$R = \frac{q_{L_I}^2 g_{L_I}^2 + q_{L_{II}}^2 f_{L_{II}}^2}{q_K^2 g_K^2}$$

with q_x the neutrino energy for capture from the xth subshell, equal to the difference between the transition energy and the binding energy of an electron in that subshell; g_x and f_x are the "large" and "small" components of the Dirac wave function for the electron.

This ratio is typically of the order of 10%.

Provided that the energy available for capture is several times the K-shell binding energy of the parent atom, the above expression also holds for forbidden transitions.

Capture from the L_{III} subshell is also possible, though since the angular momentum of this subshell is higher than that for the other L-subshells, the selection

rules are different. To allowed and first-order forbidden transitions, the contribution of L_{III} -capture is negligible. For higher-order transitions, L_{III} -capture can, in some circumstances, become important. Capture from this sub-shell is not significant in any of the isotopes considered in the present work.

The above expression shows that capture ratios depend only on the energy available for the transition and on the relative magnitude of the atomic electron wave functions. There is little dependence on the finer details of the beta-decay interaction or on nuclear parameters.

This inability of capture ratio measurements to provide information about the beta-decay interaction, together with the considerable understanding of beta-decay achieved in recent years (Konopinski, 1959) might seem to indicate that determination of capture ratios are of little interest. However such measurements supply two types of information which are rather difficult to obtain by other means.

If theoretical values of electron wave function ratios are assumed, then measurements of experimental capture ratios yield information about transition energies. Most of the energy release in capture transitions is carried

off by the neutrino and so cannot be measured directly. To determine the energy release, information from closed cycle nuclear reactions, (p,n) threshold measurements and detection of the inner bremsstrahlung associated with electron capture has to be used, and such data is rather scanty. The use of the energy dependence of capture ratios, where this is possible, is a useful tool to increase our knowledge of nuclear systematics. In those energy regions where this approach is of most use, the capture ratios are fairly rapidly varying functions of energy and are not too sensitive to the precise values of wave function ratios used, so that transition energies calculated in this manner are not liable to need revision should slight corrections to theoretical wave function ratios be required. A review of transition energies obtained in this way has been given by Robinson and Fink (1955, 1960).

In isotopes where the transition energy has been determined, the above argument can be reversed and a comparison made between theoretical and experimental values of wave function ratios. The very lack of sensitivity of capture ratios to details of nuclear structure and of the beta-decay interaction means that such comparison provides a sensitive test of our understanding

of atomic structure, virtually independent of our limited knowledge of the nucleus.

Theoretical Wave Function Ratios:

The usual method of obtaining theoretical values of electron wave functions is to assume that the atomic electrons occupy a number of separate states characterised by different quantum numbers. With this assumption, the Hamiltonian of the system is separated and analytical forms of the wave functions^{are} obtained. Separate corrections are then made for such effects as finite nuclear size and screening. In heavy atoms, relativistic effects must also be considered.

Marshak (1942) suggested the use of the wave functions obtained by solving the Dirac equation for a Coulomb potential. The use of these wave functions, given in the limit of a point charge by Bethe (1933), would automatically allow for relativistic effects, while screening effects due to the presence of the other atomic electrons could be approximated by using Slater screening constants (Slater, 1930).

Reitz (1949) computed relativistic electron wave functions, using a Thomas-Fermi-Dirac potential to

represent the mean potential of the other electrons. These results are in good agreement with those of Brysk and Rose (1955, 1958), which are considered to be the most reliable values yet prepared. Brysk and Rose computed L/K-capture ratios as a function of atomic number and of transition energy, giving full weight to corrections for finite nuclear size, using the assumption of a uniform nuclear charge distribution, to screening and to the effect of the variation of the wave functions over the nuclear volume. Their results have been used by the author throughout this thesis for comparison with experimental values.

The effect of finite nuclear size on the wave functions has also been calculated by Malcolm (1952), in the case of a nucleus in which all the charge is concentrated at the nuclear radius. Though the individual wave functions are altered, the L/K-ratio determined from this work is within a few per cent of that calculated without considering finite nuclear size effects. A similar conclusion was reached independently by Brysk and Rose.

This emphasises the lack of dependence of capture ratios on nuclear parameters referred to earlier.

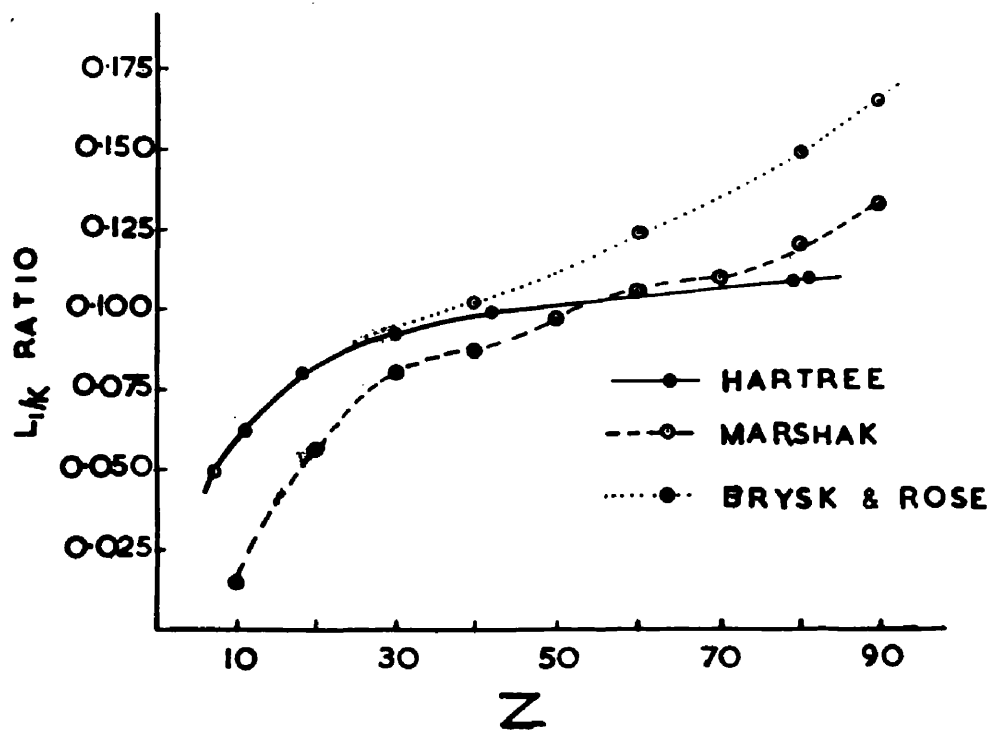


Fig.1 Theoretical L_I/K Ratios.

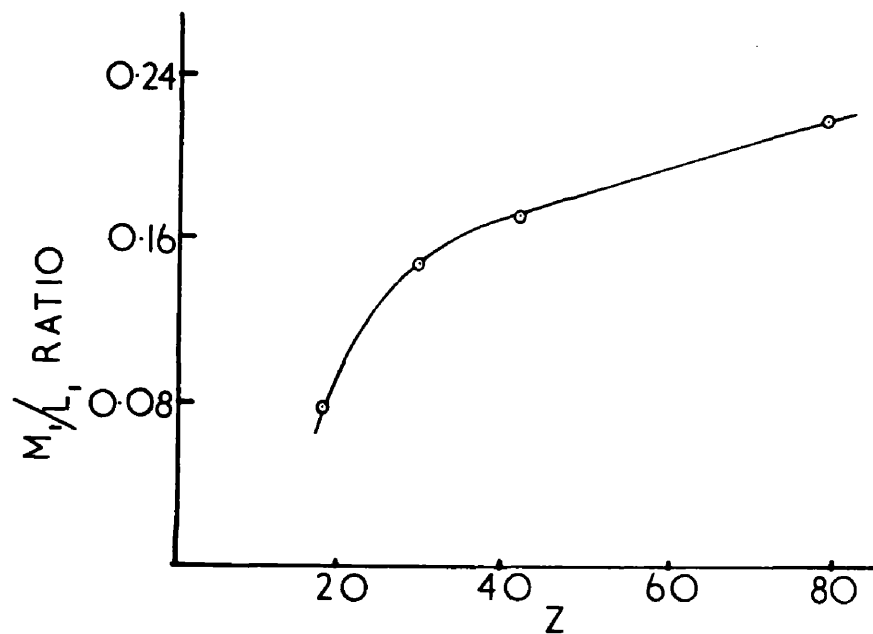


Fig.2. Theoretical M_I/L_I Ratios from Hartree wave functions.

The earliest wave functions available were the Hartree wave functions, obtained by solving the Schrödinger equation for an atomic system numerically, until the solutions are mutually consistent (Hartree 1946, Hartree 1957). Relativistic effects are neglected in this approach. Only in the case of Hg^+ have relativistic wave functions been obtained by the Hartree method (Mayers, 1957). The values obtained for these relativistic functions are quite different from the non-relativistic functions of Au^+ and Tl^+ calculated by Douglas, Hartree and Runciman (1955), showing the importance of relativistic effects at high values of atomic number.

Fig.1 shows values of L_I/K electron density ratios obtained from various sources. Analytical expressions for the Coulomb functions of Marshak and other relevant data are contained in Appendix 1. The wave functions of Reitz are not shown in Fig.1 since they yield values almost identical to those of Brysk and Rose.

The agreement among the various wave functions is rather good. At low Z values, the Hartree functions agree well with the other values, while at high Z values, where relativistic effects become important, there is good agreement between the values of Brysk and Rose and of Reitz.

Estimates of the accuracy of ratios obtained from these wave functions have been given by Brysk and Rose, who consider that their values are reliable to about 4% (Brysk, quoted in Robinson and Fink, 1960). An estimate of the accuracy can be made by calculating the atomic binding energies given by the above functions and comparing the theoretical values so obtained with the experimental values. Such a computation was carried out by Reitz (1950), who found good agreement between theoretical and experimental values, the difference being only a few per cent. Use of the ordinary Coulomb field wave functions yielded results which were considerably in error.

This computation was carried out for values of atomic number greater than about 30. From it, it is possible to conclude that theoretical capture ratios should be in reasonable agreement with experimental values in this range.

The Effect of Electron "Correlations".

Further theoretical studies of factors relating to electron capture have been made by Benoist-Gueutal (1953) and by Odier and Daudel (1956).

The former author considers in some detail the correction made to the capture ratio by the perturbation of the atomic cloud caused by the change in nuclear charge. This is shown to affect the orthogonality of the initial and final atomic states, resulting in a first order enhancement of the capture ratio for light elements. These criticisms were applied specifically to the case of Be^7 , and it was found that the L/K-capture ratio was increased by some 250% over that obtained if this contribution is neglected. This enhancement of the capture ratio falls off rapidly with increase of Z .

A further slight alteration to the ratio is caused by the alteration in the total binding energy of the atomic electrons by the change in nuclear charge. This effect, which was also considered by Benoist-Gueutal, is not important except in transitions where the energy release is small and the atomic number high.

Odiot and Daudel (1956) emphasise that the process of K-electron capture should not be regarded as the disappearance of a K-electron from the atom, but rather as a transition from a neutral atom of atomic number Z to its isobar of atomic number $Z-1$ with a vacancy in its K shell.

They discuss various methods of calculating capture ratios, with particular attention to these methods which allow for the correlations existing among the electrons. Exact calculations are extremely difficult, but from approximate calculations for Be^7 , He and A^{37} , they were able to predict that the effect of the correlations is to increase the L/K-ratio. The magnitude of the increase falls off with increase of Z, being about 3 for Be^7 , 1.25 for A^{37} and 1 for elements with atomic number greater than 20. These figures are given with reference to the values of Brysk and Rose.

Comparison with Experiment.

The conclusion reached from the above discussion is that for elements with atomic number greater than about 20, the experimental values of capture ratios should agree with the theoretical values of Brysk and Rose. For lower Z values, there is the possibility that the observed L/K-ratios will be higher, if the effects considered by Odier and Daudel are meaningful.

The rather limited experimental data available in October 1958 is summarised in Table II. In compiling this table, only the most reliable experimental results obtained for decays of known transition energy have been included,

Table II.

Isotope	L/K Ratio		Reference
	Theory	Experiment	
A^{37}	0.082	0.08 - 0.09 0.092 + 0.010 - 0.005	Pontecorvo, Kirkwood and Hanna (1949) Langevin and Radvanyi (1955)
Ge^{71}	0.106	0.128 + 0.005 - 0.003	Drever and Moljk (1957)
As^{74}	0.095	0.085 \pm 0.020	Scobie (1957)
I^{126}	0.123	0.142 + 0.005 - 0.018	Scobie and Gabathuler (1958)

It is clearly seen from Table II that agreement between theoretical and experimental values of capture ratios was considerably poorer than might have been expected.

The work described in this thesis was undertaken to provide accurate values of capture ratios in the range $15 < Z < 40$. At the upper end of this range, the theoretical values of capture ratios should not be influenced by the effects of Odiet and Daudel so that an estimate of the

accuracy of the theoretical values of Brysk and Rose could be made. Once such an estimate had been obtained, measurements at the lower end of the range would indicate whether or not the corrections of Odier and Daudel were necessary.

M-shell Capture.

During the course of the measurements, the occurrence of electron capture from the M-shell was demonstrated unambiguously in Ge^{71} and Kr^{79} and indications of its existence is obtained in Cl^{36} and A^{37} . No theoretical study of M/L branching ratios has been made, the only available source of such capture ratios being the Hartree tables. Fig.2 shows the ratio of M_I/L_I (3s/2s) electron densities obtained from the Hartree values. These have been used for comparison with experimental values.

Chapter II.

Experimental Technique.

A considerable number of different techniques have been developed for measurements of electron capture ratios in heavy isotopes. These methods have been summarized in recent reviews (Robinson and Fink, 1960; Bouchez and Depommier, 1960), and so the discussion of experimental technique given here will be confined to measurements in the range of atomic number selected for study.

Choice of Detection System.

The atom resulting from an electron capture event is excited, the excitation energy corresponding closely to one of its critical absorption energies. This energy is emitted as X-rays and Auger electrons in the atomic rearrangement which occurs within 10^{-15} seconds of capture. Since the magnitude of the excitation energy is directly related to the level from which capture has taken place, the experimental measurement of electron capture branching ratios is based on the determination of the relative number of events as a function of energy.

In the range of atomic numbers chosen for investigation, the excitation energies vary from 0.2 to 15 kev., while the fluorescence yields increase from 0.08 to 0.60. Thus

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to carry out accurate measurements of L/K-capture ratios, it is necessary to use a detector which possesses good resolution, even at energies of a few hundred electron volts, and has high detection efficiency for both Auger electrons and X-rays. The only method of carrying out measurements at such low energies is by the use of a gaseous source in a proportional counter.

Several comprehensive reviews of the characteristics of these devices have been given (e.g. West, 1953; Curran, 1955, 1958) so that discussion, where this is necessary, will be confined to points of technique which were clarified in the course of the measurements.

The use of a gaseous source eliminates the insuperable difficulties associated with the measurement of low energy Auger electrons from solid sources, while the energy per ion pair of only 30 ev. renders measurements possible down to 200 ev. The main disadvantage of using proportional counters lies in their low efficiency for the absorption of X-rays. The importance of this, which for several years delayed the accurate measurement of L/K-capture ratios in the range of atomic number considered here, is as follows.

After a K_{α} X-ray has been emitted, the atom is left ionised at the L_{II} or L_{III} level, and since the partial

L-shell fluorescence yields are very small, this energy is released as Auger electrons. If the K_{α} X-ray is absorbed in the counter, the total energy detected corresponds to the K-binding energy of the daughter atom and the event is registered in the K-peak. However, if the X-ray escapes from the sensitive volume of the counter, the event is counted in the L-capture peak and the energy resolution of the counter is not sufficient to distinguish between genuine L-capture events, which are almost 100% from the L_I subshell, and these apparent L_{II} - or L_{III} - capture events arising from K X-ray escape. Thus, there is a systematic error in the measured L/K-ratio and corrections are required to obtain the true value.

The magnitude of the error depends on the fluorescence yield of the atom and on the relative absorption efficiency of the counter for X-rays. Since there are usually ten times as many K-events as L-events, any miscalculation in the amount of K X-ray escape is magnified in the correction to the observed capture ratio. While the flux of X-rays escaping from standard geometries can be estimated with some confidence, values of fluorescence yields are not accurately known and techniques which depend on the use of these values are unreliable.

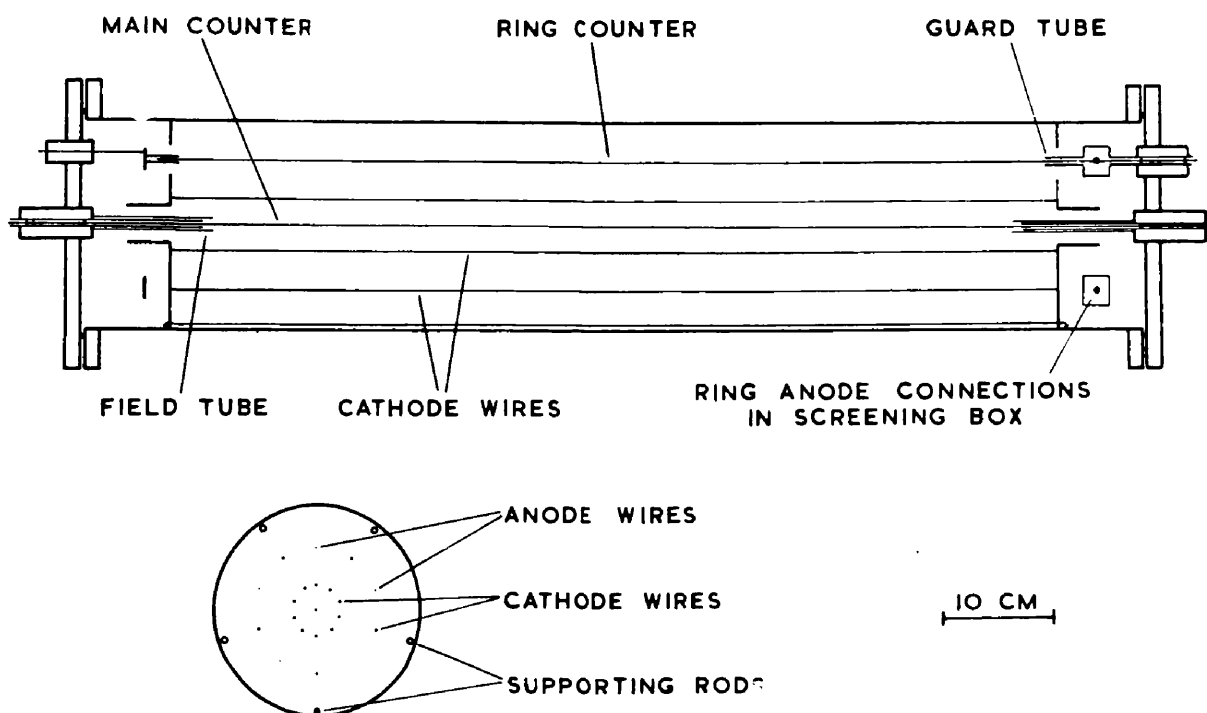


Fig.3. The wall-less counter used in measurements on Ge^{71} and Kr^{79} .

Two methods have been used to overcome this limitation.

In the first, used for measurements on A^{37} by Pontecorvo, Kirkwood and Hanna (1949) and by Langevin and Radvanyi (1955), xenon was used as filling gas in a conventional counter. Because of its large Z value, this filling possessed high detection efficiency for the 2.6 kev. K X-rays of chlorine and the escape of X-rays amounted only to a few per cent.

This technique has not been used for other isotopes, since for higher energy X-rays, the pressure of xenon required becomes too great.

The other technique is the ingenious "wall-less" counter method first described by Drever and Moljk (1957).

The Wall-less Counter Technique.

The principle of the wall-less counter may be understood by reference to Fig.3. The device consists of two proportional counters, one entirely enclosed within the other, connected in anti-coincidence. The common cathode of the counters is a circle of wires connected directly to the case. The outer counter, referred to as the ring system, comprises a number of separate counters defined by the common cathode and by the outer circle of wires, which are alternately cathodes and anodes. With a gaseous

source and a pressure of filling gas such that the ring system is several half-thicknesses deep to X-rays from the central counter, the spectrum of pulses from the latter in anti-coincidence with pulses from the ring is measured. The relative intensities of the observed K- and L-event peaks are then very nearly the true K- and L-counting rates.

The number of events in the L-peak is close to the number of L-capture events, since, in those cases where a K_{α} X-ray escapes from the inner counter, it is absorbed in the ring and so operates the anti-coincidence mechanism. Hence, only genuine L-capture events are recorded.

The number of K-events is also close to the true number, though in this case the peak is made up of two components. The first arises from K-Auger events and from those K fluorescent events which are detected in the central counter. These events give rise to pulses corresponding to the K-absorption energy. The other component arises from K X-rays which are produced in the ring system and detected in the central counter. These X-rays are detected provided that pulses in the ring corresponding to the L-absorption energy do not operate the anti-coincidence circuit. To a good approximation, the inner counter is surrounded by an infinite medium containing a uniform source distribution and so equal

numbers of X-rays pass from one counter into the other. The number of events recorded in the K-peak is the number of K-events in the central counter. Though the two components of the peak differ in energy by several per cent, the resolution is such that both components are integrated into one peak.

It is clear from the above description that the dependence of the observed capture ratio on the fluorescence yield is all but eliminated in this technique.

Some minor corrections must however be made to the observed capture ratio, which is obtained by comparing the relative area of K- and L-peaks.

The ring is, in practice, only a finite depth, so that there are always some X-rays which pass from the central counter through the ring and into the wall. These X-rays give rise to apparent L-events, as do those X-rays which leave the central counter and are absorbed in the wires forming the common cathode. Both these effects also make the observed K-counting rate slightly smaller than the true one.

A third correction, which does not affect the K-counting rate if the counter is fitted with field tubes,

arises from the escape of X-rays out of the unshielded ends of the counter.

If these corrections are denoted by P_1 , P_2 and P_3 respectively, the fluorescence yield by F and the fraction of K_α X-rays in the K series by k , then the true L/K-capture ratio R is related to the observed ratio R' by

$$R = R' \left[1 - (P_1 + P_2)F \right] - (P_1 + P_2 + P_3)Fk \quad (4)$$

Because of the unusual geometry of the counter, the calculation of these corrections cannot be made rigorously. The corrections can be made small by suitable choice of the counter dimensions and pressure of filling gas. Approximate calculations can then be made by use of formulae developed for the fluxes of X-rays out of standard source geometries, given by Hammersley (1952), Price, Horton and Spinney (1957) and Glasstone (1956).

In measurements in which M-capture is observed, corrections for apparent M-capture events arising from the escape of K_β X-rays can be made in a similar manner.

Since corrections for escape of X-rays can be minimised in a fairly simple manner, wall-less counter techniques have been used throughout the work to be described.

The main practical limitation to the use of these devices is the necessity for obtaining the source in gaseous form. For many of the elements on which measurements would be desirable, it is impossible to obtain suitable liquids or gases satisfying the rather stringent conditions necessary for use in a counter.

The compound must not possess a high electron attachment coefficient for the range of E/p values encountered and it must be chemically stable. In particular, it must not be sensitive to the presence of small traces of common reagents such as are usually found on the walls of counters. Further difficulty arises because the avalanche process produces ultra-violet photons and if the substance is photo-sensitive, it is gradually decomposed. These considerations are illustrated by the work carried out by the author and Mr K.W.D. Ledingham on Zn^{65} in an attempt to measure capture ratios in that isotope. The presence of slight traces of alcohol on the counter walls caused the zinc di-ethyl used as source carrier to decompose. The work had to be abandoned.

Two different counters were used in the measurements. Details of these counters are given in Appendix 2.

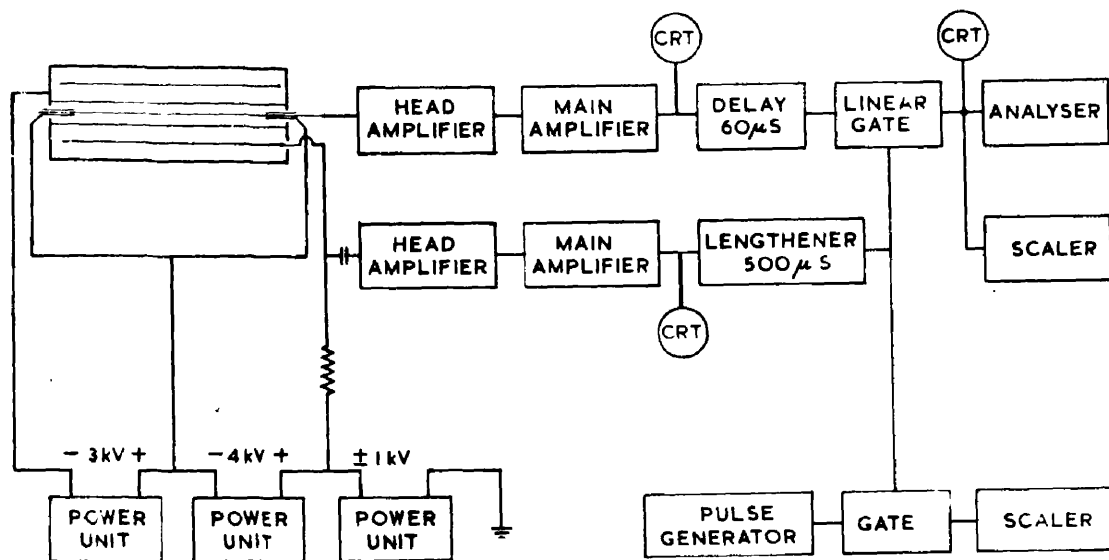


Fig.4. Initial form of electronics.

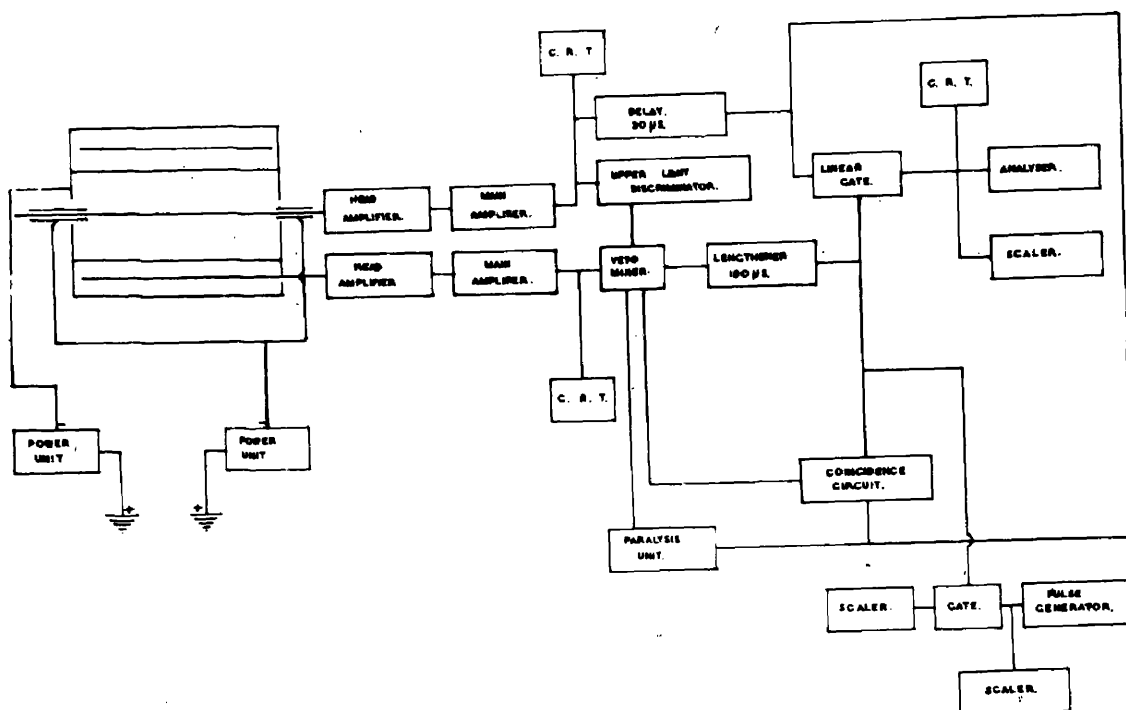


Fig.5. Final form of electronics.

Electronic Circuits.

Basically the use of a wall-less counter technique requires the use of standard electronic anti-coincidence circuits. The circuits initially employed in the present work are those used by Drever and Holjk (1957), which are shown in Fig.4. These circuits have been described by Drever (1958).

As experience was gained in the use of wall-less counters over a wide range of experimental conditions, various units were added to this basic form, resulting finally in the layout shown in Fig.5. Reference is made to the functions of these added units at the points where they were introduced.

A brief account of the details of the new units added is given in Appendix 3.

Chapter 3.

Electron Capture Ratios in Germanium-71.

Introduction:

The most suitable isotopes for accurate measurements of orbital electron capture ratios are those in which the decay proceeds from nuclear ground state to ground state solely by a capture transition. Not only is the measurement simplified, but the analysis of the results is not complicated by the need to consider effects arising from competing nuclear processes, such as internal conversion. Among such isotopes occurring in the range selected for study are Λ^{37} , Fe^{55} and Ge^{71} . Measurements on both Λ^{37} and Ge^{71} are included in Table II.

An early measurement of the L/K-ratio in Ge^{71} was reported by Langevin (1954, 1956), who used a source of germanium hydride in a proportional counter. The filling gas was propane so that most of the X-rays escaped detection and the L/K-ratio was deduced from the K and L Auger electron intensities. This measurement depends critically on the value used for the fluorescence yield of gallium. The L/K-ratio varies from 0.30 if the figure

of 0.45 is used for the fluorescence yield, as suggested by a semi-empirical formula of Burhop (1952), to 0.14, using the experimental value of 0.53 reported by Drever and Moljk (1957).

The best experimental value of the L/K-ratio in Ge^{71} was obtained by Drever and Moljk (1957). Their value of 0.128 ± 0.005
 $- 0.003$ was about 20% higher than the value of 0.106 calculated from the data of Brysk and Rose, and the difference between theory and experiment was greater than might have been expected in this region. This work of Drever and Moljk not only pioneered the application of wall-less counter methods to studies of electron capture ratios, but was also the first measurement in which proportional counters were employed at pressures of several atmospheres to examine events in the 1 kev energy region. Other workers with high pressure counters have measured spectra at considerably higher energies so that lower values of gas gain were used (see, for instance, West et al., 1955).

Since the publication of this result, Drever had acquired considerably more experience in the use of proportional counters at high pressures, and had studied several points of technique in some detail. In particular,

it was found that the resolution of peaks at low energies appeared to improve as the diameter of the anode wire was decreased.

This observation was investigated by the author and it was found that the effect was caused by non-uniformity of the anode wires producing local variations in field strength, and hence fluctuations in gas gain, both along and round the wire. The low fields associated with thin wires permit the initial electrons formed to diffuse as they are pulled towards the wire, so that the effective shape seen is an average over the whole wire. With thick wires and higher fields, the diffusion is reduced and the local shape of the wire becomes correspondingly more important.

That the non-uniformity of the wire was responsible for the deterioration in resolution was confirmed by some spectra taken in which platinum wire of high uniformity was used for the anodes. The peaks obtained with this wire were about a factor of 2 better in resolution than those obtained with tungsten wire of the same diameter.

Similar effects of variation in gas gain along the length of a wire have been reported by West et al.

Measurements on Ge^{71} are important because they provide one of the critical tests of theory. The possibility of a systematic error of technique in the earlier work of Drever and Moljk led to the decision to repeat their measurement.

The absence of both particle and gamma emission in the decay of Ge^{71} makes this isotope a convenient source for the study of internal bremsstrahlung, and such investigations have been reported by several groups. Saraf et al. (1953) and Saraf (1954) found good agreement with the theoretical spectrum shape down to 100 kev. Langevin (1954) found a low energy component in the spectrum and considered that an isomeric state of Ge^{71} existed, its half-life being considerably greater than that of Ge^{71} itself. Bisi et al. (1955) made the most thorough study and showed that when well-purified sources were used, the spectrum obtained was simple and agreed with the theoretical distribution down to 50 kev. From these measurements, the transition energy is 233 kev and the transition is allowed, with $\log ft = 4.6$. The half-life is 12.5 ± 0.1 days (Bisi et al.).

Source Preparation:

Ge^{71} was prepared by neutron irradiation of germanium oxide at Harwell and was stored long enough for the 12-hour activity of Ge^{77} also produced to decay.

The source was converted into germanium hydride by reaction with nascent hydrogen released in an aqueous solution of the oxide by the action of sodium amalgam. Water vapour was removed by freezing and excess hydrogen pumped off.

Sufficient germanium hydride to give a counting rate of some 35,000 counts per minute in the total volume was put into the high-pressure counter described in Appendix 2 and argon, to a total pressure of 6 atmospheres, added. Methane to a partial pressure of 15 cm. provided the quenching. With this filling, the total escape corrections amount to about 7%.

During most of the measurements, the electronic circuit of Fig.4 was used, the dead-time of the gate being 1 milli-second and the delay time 60 micro-seconds.

At high pressures, there is a large background counting rate from cosmic rays and natural activities. To give the best possible ratio of source to background,

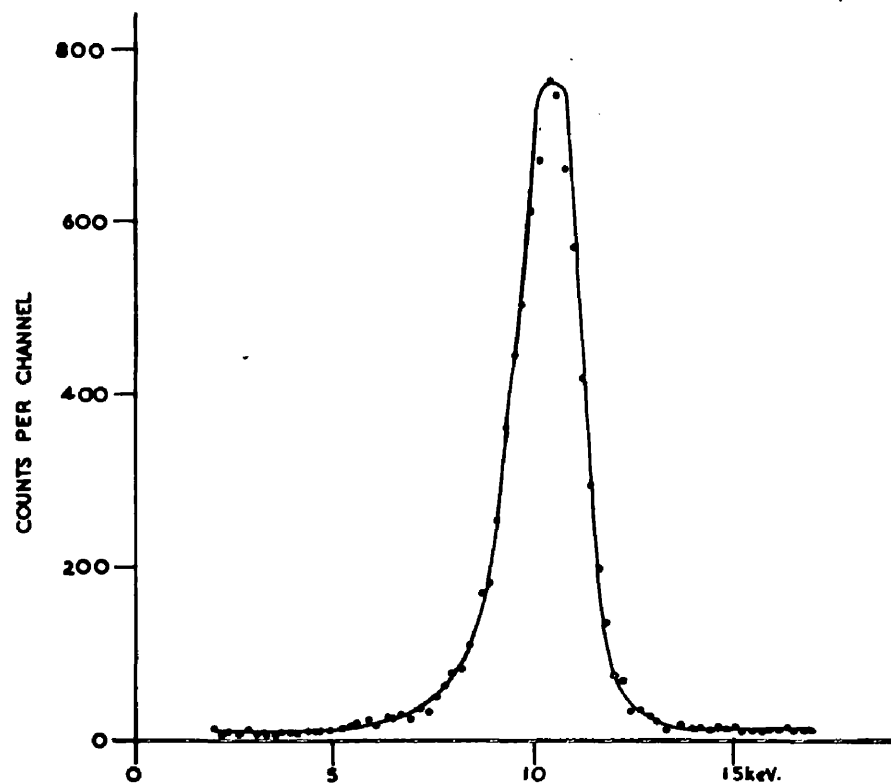


Fig.6. Ge^{71} K-peak in the central counter in anti-coincidence with events in the ring.

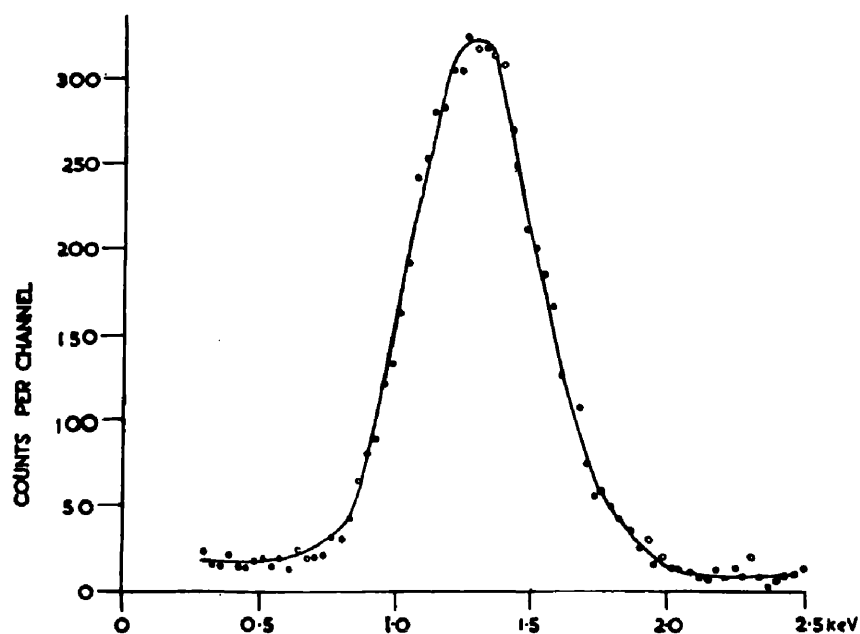


Fig.7. Ge^{71} L-peak in the central counter in anti-coincidence with events in the ring.

Ge^{71} L

the counter was shielded by a lead castle, the minimum wall thickness being 2 inches.

L/K Ratio Measurements.

With the experimental conditions outlined above, the pulse spectra in the region of the 10.3 kev K-peak and the 1.3 kev L-peak in the central counter, in anti-coincidence with events in the ring counter, were analysed. The gas gain was kept constant throughout the measurements, the change in energy range being made by using different amplifier gains. Typical K- and L-peaks obtained under these conditions are shown in Figs.6 and 7.

The mode of operation of the technique requires that the anti-coincidence circuit is able to discriminate between K- and L-events in the ring. A spectrum of events in the ring is given in Fig.13 for the similar measurements carried out on krypton-79, the energies of K- and L-events being about the same for the two isotopes.

It has already been shown that the bias level in the ring must lie between the K- and L-peaks. As a check on the functioning of the apparatus, runs were taken with various values of bias level.

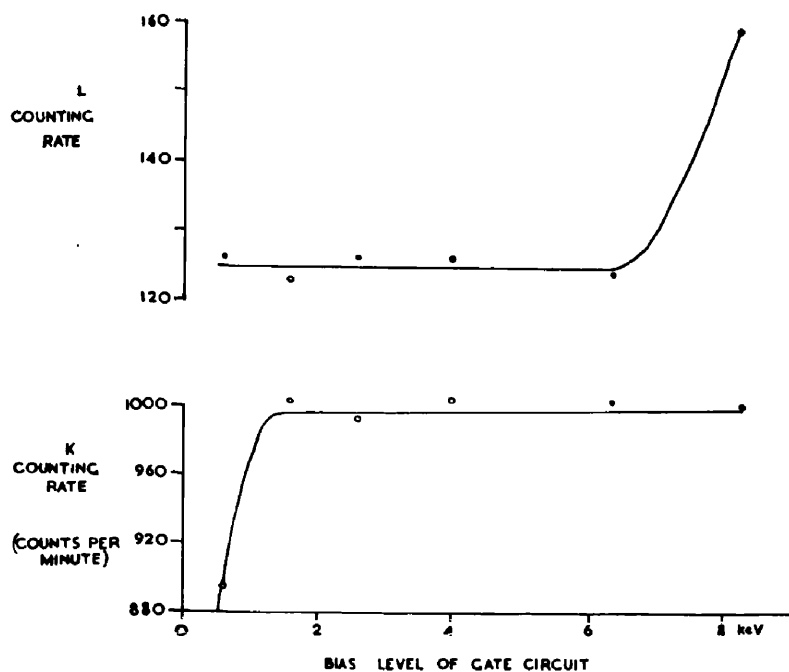
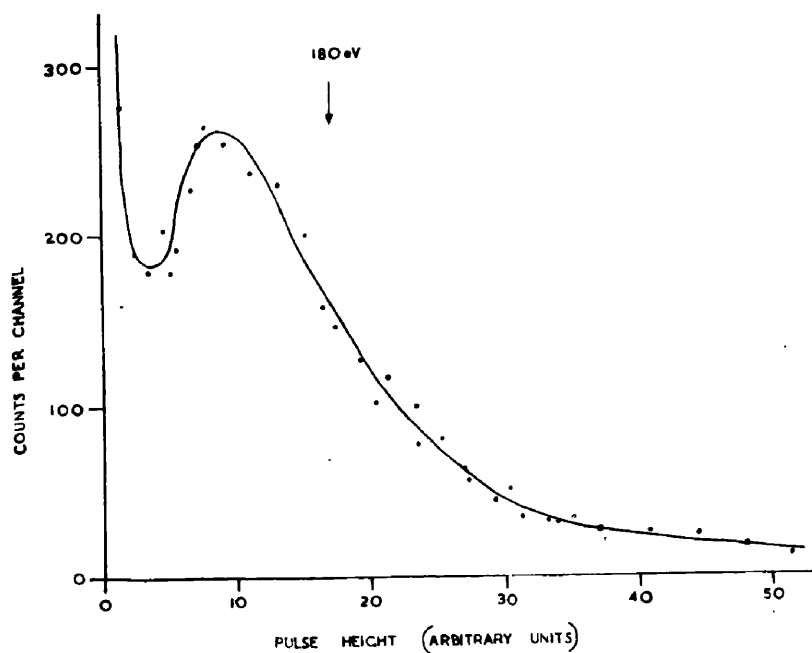


Fig.8. Variation of counting rate with ring bias.



Ge⁷¹ M

Fig.9. Ge⁷¹ M-peak in the central counter in anti-coincidence with events in the ring.

If the bias level is too high, then some of the K-events in the ring counter will not close the gate, and so a number of apparent L-events in the central counter will be recorded, while if the bias level is too low, the gate will be closed by some of the L-events in the ring, and hence some of the K-events passing from the ring into the central counter will not be counted. In both cases, the observed L/K-ratio will be too high. Between these two extreme positions, the ratio should be constant. Graphs of the variation of K- and L-counting rate with bias setting are given with Fig.8. The expected variation of K- and L-counting rates with bias level occurs.

Most of the measurements were carried out at 6 atmospheres pressure. However, since the escape corrections can only be calculated in an approximate manner, it was decided to check on the validity of the calculations by taking a set of measurements at 12 atmospheres pressure, at which pressure the corrections are approximately halved.

The results obtained from these measurements are summarized in Table III.

Table III.

Pressure (Atmospheres)	Escape Corrections	Number of measurements	Average result	Corrected result
6	$P_1 = 7 \times 10^{-4}$ $P_2 = 7 \times 10^{-3}$ $P_3 = 7 \times 10^{-3}$	5	0.1235	0.1167
12	P_1 negligible $P_2 = 3.5 \times 10^{-3}$ $P_3 = 3.5 \times 10^{-3}$	3	0.114	0.1156

$$F = 0.50, \quad k = 0.86$$

The symbols here have the same meanings as in (4) in Chapter 2.

Corrections have been made in calculating the above ratios for the dead-time introduced by the gate circuit and by the Hutchinson-Scarrott analyser used in the measurements. These corrections are less than 1% here. The statistical error in the individual measurements is about the same size.

An early set of measurements at 6 atmospheres has not been included in the above table because the gain in the ring system drifted while the measurements were being performed. The results of these measurements are in good

agreement with the value given above. The drift in the ring was corrected before the final measurements were carried out.

One small correction, which is needed only when the energy of the X-rays being measured is greater than the K-absorption energy of the filling gas, remains to be made. K X-rays from the ring pass into the central counter and are absorbed there by the photo-electric effect in the K-shell of argon atoms. If an argon atom de-excites by X-ray emission, then it is possible for the X-ray to pass back into the ring counter and be absorbed. Thus the total energy detected in the ring is about 4 kev. This is usually sufficient to operate the anti-coincidence circuit and so leads to a loss of genuine K-events.

It was calculated that this correction amounted to about 1% at 6 atmospheres and was negligible at higher pressures.

After all the corrections had been made, the L/K-electron capture ratio in germanium-71 was determined to be 0.116 ± 0.005 , the error arising mainly from the determination of the shape of the peaks.

Though this value is lower than the earlier result of Drever and Holjk obtained by the same technique, it

is still higher than the value of 0.106 calculated from Brysk and Rose. The difference between the results of the two experimental measurements can be explained by the considerable improvement in energy resolution in the present measurement, brought about by the use of thin anode wires.

The question of whether or not the small difference still remaining between theory and experiment is significant or whether it is due to some unsuspected experimental limitation, such as was present in the earlier measurement, is discussed at the end of the next chapter.

Some time after this work was completed, a scintillation counter measurement of the L/K-ratio in Ge^{71} was reported by Rehfuss and Crasemann (1959). Their value of 0.09 ± 0.05 is in agreement with the value obtained above.

M/L-Capture Ratio Measurements.

Electron capture from the M-shell is also possible. At this period, only one definite observation of M-capture had been reported (in Np^{235} by Gindler et al., 1958), although its occurrence had been inferred in a number of

other cases (for instance, Huizenga and Stevens, 1954; Magnusson et al., 1957).

It was decided to examine the low energy spectrum of Ge^{71} to determine whether or not it was possible to detect events arising from M-capture. The M_I -absorption energy of gallium has been estimated to be 182 ev. (Hill et al., 1952). The only other study of events at such a low energy with proportional counters is that by Scobie and Lewis (1957) who were able to detect K-capture events in C^{11} , the energy release being about the same magnitude.

After the measurement of the L/K-ratio had been completed, the gas gain in the central counter was increased by about a factor of 10, the bias potential on the ring system being changed to keep the effective potential difference across the ring counters unaltered.

The two sets of counters are not independent. Variation of the potential difference across either counter alters the position of the virtual cathode and so affects the pulse height in the other. For this reason, it is essential to make comparison of the areas of, for instance, K- and L-peaks, under identical conditions, the only adjustable parameter being the amplifier gain.

This uncertainty in the exact diameter of the counter is reflected in the calculation of the escape corrections, but since these are small, little error is produced in the final result.

The first measurements in the energy region below 1 kev indicated the presence of a peak at approximately the correct energy, but alterations had to be made to the gate circuit before reliable measurements could be carried out.

The difficulty arose from the presence of about 50 times as many K-events as M-events. Because these K-events produced pulses of some 50 times the pulse height of the M-events, the gate circuit tended to overload, even although all the units were designed with D.C. coupling wherever possible to minimise the degree of overload. The upper limit discriminator of Fig.5 was added to the circuit at this point to remove the difficulties associated with the presence of the K-pulses. This unit is such that all signal pulses larger than a certain height, which is usually about 5 volts greater than the maximum accepted by the analyser, operate the gate veto and so close the gate for the duration of the pulse together with the

gate dead-time. This gives the gate time to recover from any overload induced. Further improvement in performance was achieved by lengthening the gate dead-time.

Under these conditions, the spectrum in Fig.9 was obtained. This contains three separate measurements at different amplifier gains which have been fitted together with due allowance for channel width, which was calibrated under the conditions of the experiment with a pulse generator, and for gate and kicksorter dead-times.

The occurrence of a peak at about the energy expected is clearly seen. Though apparent M-events can arise from the escape of K_{β} X-rays, the observed intensity of the peak is 10 times greater than can be accounted for by this mechanism alone and the existence of M-electron capture in germanium is thus confirmed. The intensity of the peak relative to the L-peak, is 0.18 and after correction for the escape of K X-rays, the M/L-ratio in Ge^{71} was determined to be 0.16 ± 0.08 . The ratio derived from the Hartree wave functions of Fig.2 is 0.153, in good agreement with the above result. The large error

is due to the difficulty of determining the shape of the peak because of the rapidly rising background.

The peak occurs at a slightly lower energy than the 180 ev expected. This difference is not considered to be important.

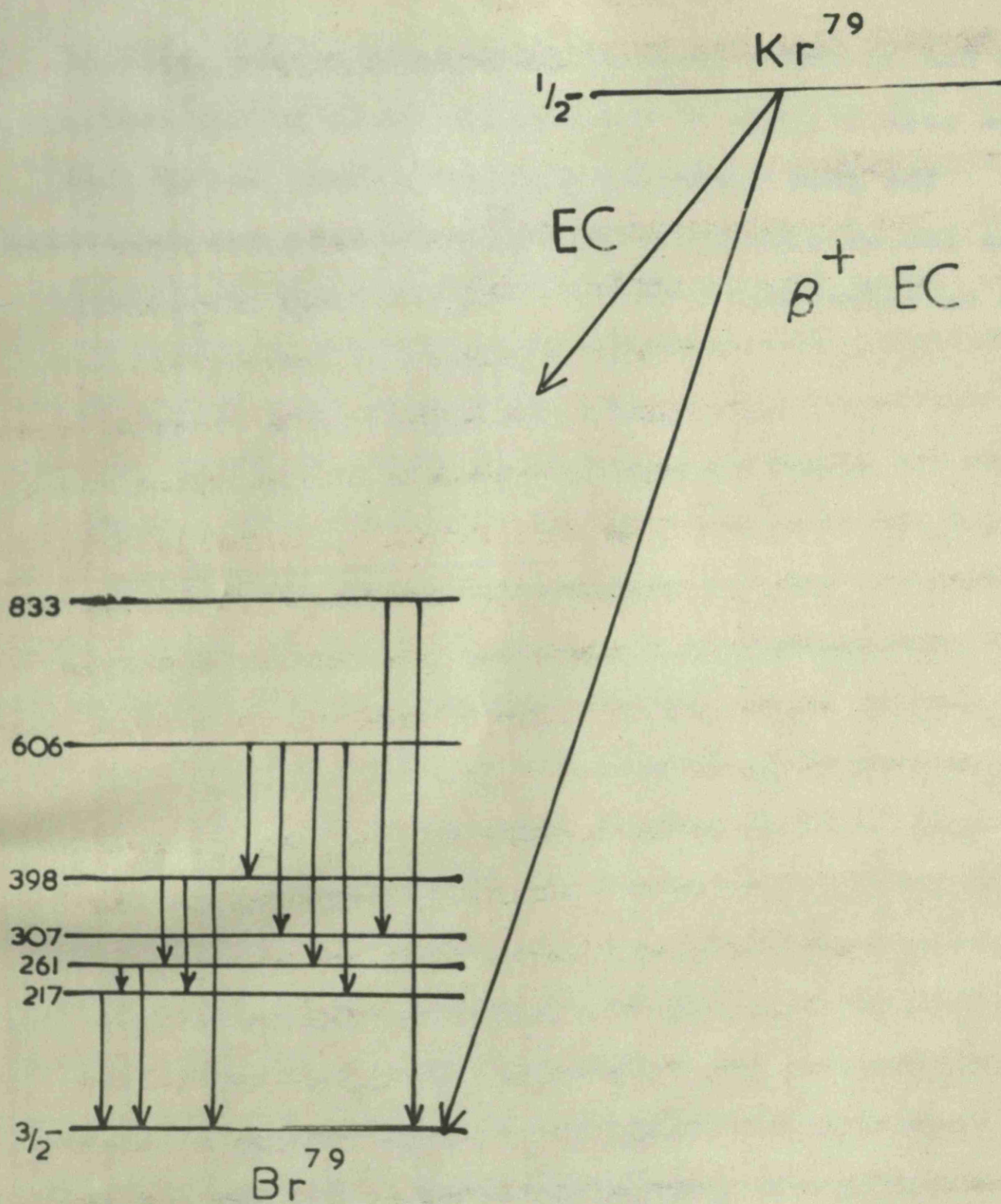


Fig.10. The decay scheme of krypton-79.

Chapter 4.

Orbital Electron Capture Ratios in Krypton-79.

Introduction:

The measurements on Ge^{71} reported in the previous chapter appear to indicate the existence of a small discrepancy between experimental and theoretical values of L/K-capture ratios in the intermediate range of atomic number. Since it was expected that the calculations of Brysk and Rose would be rather more accurate in this region than the agreement with experiment suggests, it seemed desirable to measure capture ratios in another isotope of about the same atomic number, to confirm that the discrepancy was real.

The most suitable isotope in the neighbourhood of germanium-71, for such a study, is krypton-79. The decay scheme, shown in Fig.10, is based principally on the work of Thulin et al. (1954) and Thulin (1955). Though the complete details of the energy levels are not free from ambiguity, the smallest energy available for any of the capture transitions is 800 kev and the spread produced in capture ratios by the energy dependent factor is less than 1%. The half-life is 34.5 hours (Radvanyi, 1952). Though this is rather short, it is

compensated for by the fact that krypton is naturally gaseous, so that no delay is imposed by the need for chemical preparations. The decay is allowed with $\log ft = 5.4$. About 5% of the decay proceeds by positron emission.

Effects of Internal Conversion:

With a decay scheme as complex as that of krypton-79, consideration must be given to the effect of internal conversion on the observed capture ratio. The long resolving time of the counter makes it an integrating device, so that in those disintegrations in which electron capture is followed by internal conversion, the energy observed in the counter is different from that corresponding to a capture event and the event is not counted in the relevant capture peak. Thus a genuine capture event is lost and corrections have to be made for this effect. This correction becomes negligible either if the internal conversion ratio is approximately the same as the capture ratio, or if the absolute internal conversion coefficient is small.

Internal conversion coefficients of the more abundant gamma rays of Kr^{79} have been measured by

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Bergström (1951, 1952), and it is found that both the above conditions are fairly well satisfied, so that the effects of internal conversion on the capture ratio can be neglected.

Earlier Measurements:

Values of the L/K-ratio in Kr^{79} had already been obtained in two different ways. Radvanyi (1952) used Kr^{79} in a cloud chamber and made a measurement of the K- and L-Auger electron intensities in a small region, obtaining the value of 0.27 ± 0.09 . In the other measurement, Langevin and Radvanyi (1954, 1956) used the technique of Langevin (1954) discussed in connection with Ge^{71} . The result determined in this work was 0.257 ± 0.030 . Both these methods suffer from the disadvantage of depending strongly on the value of the fluorescence yield of bromine. The value of 0.586 for this quantity used in the second measurement quoted above is based on the work of Laberrigue-Frolow et al. (1956). If instead the value of 0.60 is used, the L/K ratio becomes 0.16, in better agreement with the value of 0.101 calculated from the graphs of Brysk and Rose.

There is seen to be considerable disagreement between the above values and the theoretical value.

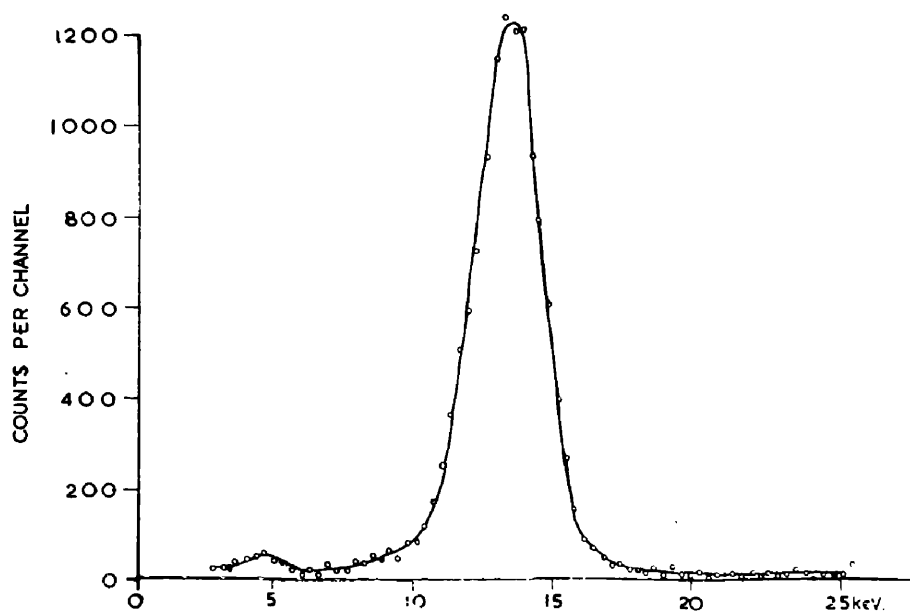


Fig.11. Kr^{79} K-peak in the central counter in anti-coincidence with events in the ring.

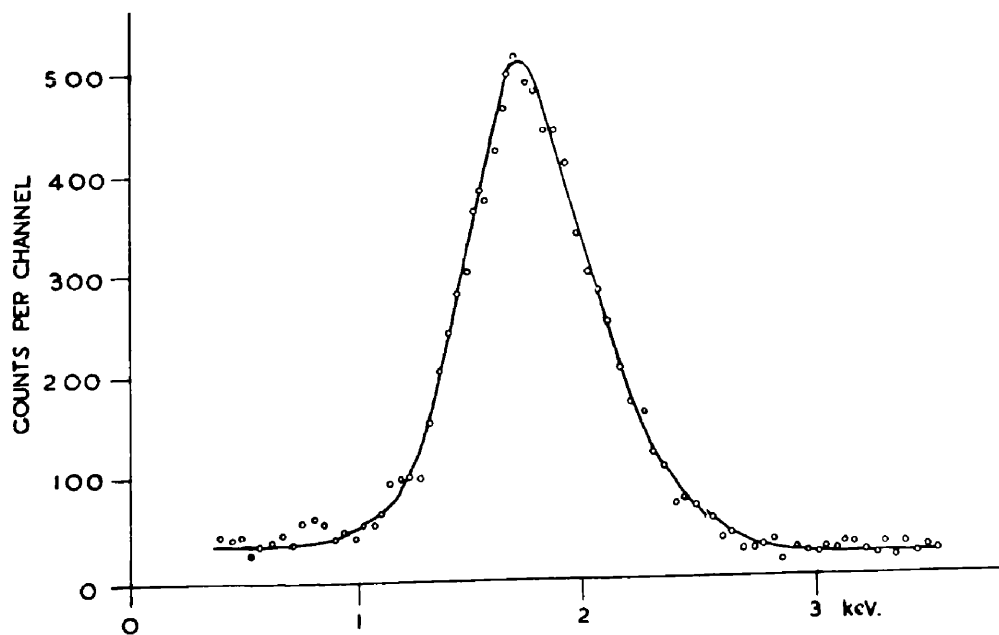


Fig.12. Kr^{79} L-peak in the central counter in anti-coincidence with events in the ring.

The measurements in the present chapter were undertaken partly to clear up this disagreement by measuring the value with a technique not so dependent on fluorescence yield, and partly to provide another reliable measurement of an L/K-ratio to facilitate critical comparison of theory and experiment in this region.

Experimental Details.

Kr⁷⁹ was prepared by (n,γ) reaction at Harwell.

The same counter was used as in the work on Ge⁷¹.

The filling was 15 cms. of methane and 12 atmospheres of argon, and was such that the escape corrections were unaltered from the previous measurement. After filling, the gases were left for some 12 hours to ensure thorough mixing.

The experimental conditions were otherwise unaltered.

L/K Measurement.

Figs. 11 and 12 show typical K- and L-peaks of Kr⁷⁹ from the central counter in anti-coincidence with events in the ring. The slight peak at about 5 kev. in Figs. 11 and 13 was attributed to the presence in the source of an electron-capturing isotope of xenon. The presence of this impurity did not affect the results.

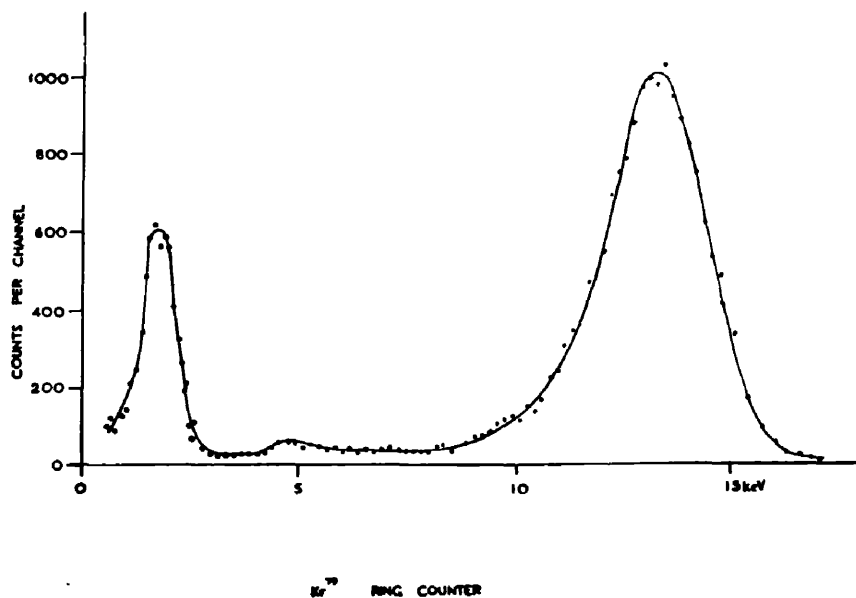


Fig.13. Ring spectrum of K- and L-capture events in Kr^{79} .

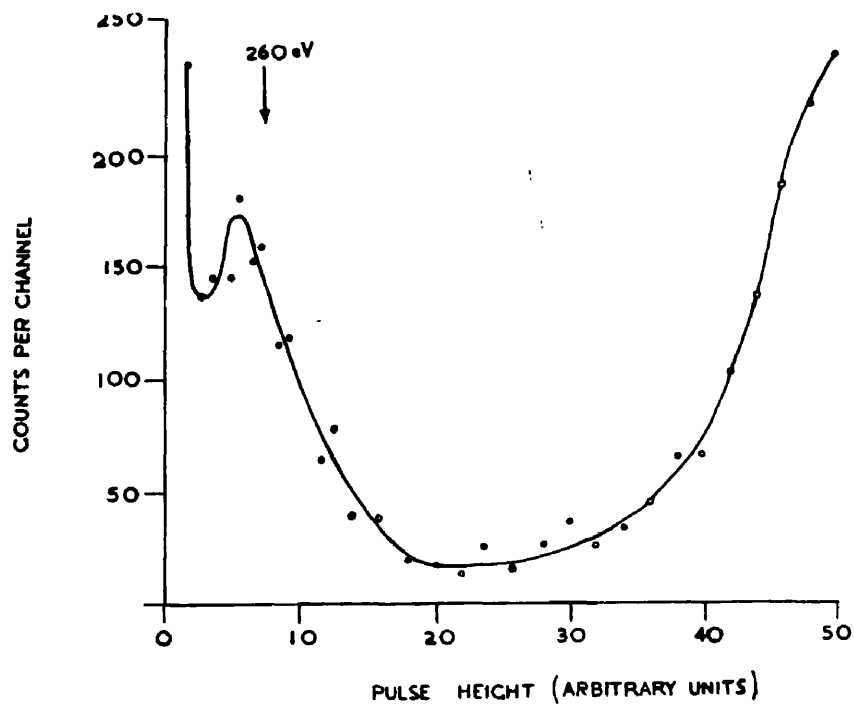


Fig.14. Kr^{79} M-peak in the central counter in anti-coincidence with events in the ring.

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The peaks obtained in these measurements were not appreciably worse in peak to valley ratio than the germanium-71 peaks, showing that background events arising from the positron component of the source were almost entirely eliminated.

Fig.13 shows a spectrum in the ring. The clear distinction between K- and L-events necessary for the proper functioning of the technique is present.

Two sets of runs were taken. In the first set, small variations in dead-time were induced by the occurrence of grid current in the pulse lengthener. Steps were taken to remove this before the more reliable second set of measurements was made.

As before, measurements were carried out with different values of bias level.

Because of the short half-life, the values obtained had to be corrected for decay, over and above the other corrections for dead-time mentioned in connection with the results for germanium-71. This was carried out by normalizing individual sets of runs to a fixed time.

After correction for escape had been made, the L/K-ratio in Krypton-79 was determined to be 0.108 ± 0.005 .

The values of $F = 0.60$ and $k = 0.83$, in the notation of equation (4), have been used in calculating this value from the experimental figure of 0.116.

M/L-Capture Ratio.

Measurements were undertaken to confirm that M-capture existed. The experimental technique was the same as above, the only difference being that the gate dead-time had to be increased to about 5 milli-seconds, so that the gate was only open about 25% of the time. With shorter dead-times, it was not possible to obtain M-peaks.

Fig.14 shows the peak obtained. The individual runs making up this figure have been fitted together with full allowance for decay, dead-time and channel width.

The value obtained for the M/L-ratio after correction for the escape of K X-rays is 0.16 ± 0.08 , in agreement with the value of 0.16 obtained from Fig.2. The peak is once again rather lower in energy than was expected.

Discussion of the Results of Chapters 3 and 4.

The values of L/K-ratio obtained in these two measurements are both slightly higher than predicted by theory. Since the earlier measurement on Ge⁷¹ by Drever and Moljk (1957) had contained an unsuspected technical error, the whole principle of the technique was subjected to careful scrutiny to make quite certain that no possible source of systematic error had been overlooked.

For instance, an estimate was made of the probability for the path length of the K-Auger electrons detected being sufficiently long for some of the tracks to begin in one counter and end in the other. If this happened, then some K-events would not be counted in the K-peaks and the observed L/K-ratio would be too high, as is actually the case. Another effect considered was whether there was at the common cathode a dead space deep enough for events occurring there to straggle into the wire and not develop their proper pulse height. These effects were found to be negligible and it was concluded that there were no fundamental systematic errors in the technique.

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Analysis was made of the numerical results obtained to see if the slight tails present on the peaks should be included in the counts ascribed to the corresponding events. To some extent this is checked on by the runs in which the variation of L-counting rate with ring bias is measured, since the counting rate would be slightly altered if these tails belonged to the K-peak. Various methods were used for evaluating the number of counts in the peaks. No dependence of the calculated L/K-ratios on the method used to evaluate the number of events in the peak or on the bias level of the anti-coincidence circuit was observed, within the energy region where the L/K-ratio is expected to be independent of the ring trigger level.

Natural background was measured separately and was found to be too small in the energy region of interest to account for the tails on the peaks.

In another measurement, a beta source (Kr^{85}) was put into the counter and the resulting spectrum examined under the conditions of the Kr^{79} runs to see if the tails observed in that measurement arose from the positron component of the source. This is not an exact test

because Kr^{85} emits electrons, but since there are more low-energy electrons than positrons, the tails, if due to this cause, would be exaggerated. The effect of the presence of the source was negligible. No amount of the source compatible with the observed counting rates could produce spectra of the required intensity and shape.

These measurements provide a check on the operation of the counter, but there still remains the possibility that, with the particular electronic circuits and the rather generous length of pulses (some 50 micro-seconds long) used in the measurements, spectrum distortion could be produced by electronic means. A run was carried out in which the counting of a K-capture peak was simulated by using a suitable pulse generator, with pulses from a counter gating this spectrum at the correct rate. All the pulses were passed through the same electronic circuits as in the actual measurements, in this way duplicating the conditions of the experiment. The spectrum obtained was examined for electronic effects; none were found. Despite their complexity, the electronic units were not responsible for spectrum distortion.

It can only be concluded from the above measurements that the discrepancy between theory and experiment, though small, is none the less genuine and that the results of Brysk and Rose are a few per cent too low in this range of atomic number.

In 1956 when Odier and Daudel discussed the influence of electron "correlations" on capture ratios, the discrepancy between theory and experiment was about a factor of two, and they concluded that since the relative magnitude of the correlations fall off rapidly with atomic number, an increase in theoretical capture ratio sufficient to make theory and experiment agree could not arise from this effect. Since that time, the agreement between theory and experiment has been much improved and it is possible that the small difference now remaining can be accounted for in this way. If this interpretation, that the "correlation" effect of Odier and Daudel must be considered, is correct, then it follows that measurements on isotopes of small atomic number should give values of capture ratios in considerable disagreement with the theoretical results of Brysk and Rose. For this reason, attention was directed to

the measurement of L/K-ratios in light isotopes. Such measurements form the basis of the next two chapters.

Electronic Modifications.

Various improvements in the gate circuit were suggested by experience gained in the above measurements.

The use of a long dead-time after both signal and veto pulses, as in the M-capture measurements in Kr⁷⁹, is unnecessary, since it is only after signal pulses that precautions to avoid overloading effects are required. A unit was incorporated in the circuit such that after every signal pulse a pre-determined dead-time was inserted. This is the paralysis unit of Fig.5. The combination of the upper limit discriminator with this paralysis unit eliminates completely the possibility of signal pulses being wrongly analyzed because of overloading either of the amplifier or of the kicksorter. The upper limit closes the gate before the pulse arrives and the paralysis unit keeps it shut for a period sufficiently long to allow all units to return to their quiescent condition.

The other unit added to complete the transformation of the circuits from Fig.4 to Fig.5 was a coincidence unit. If a genuine signal pulse arrives at the gating valve as it is about to open at the end of the gating

pulse associated with a previous signal, the gate remains shut. This unit removes the last way in which a pulse can be distorted by the electronic circuits. At the same time, it leads to a slight loss of genuine signal pulses and a small correction has to be made to the observed dead-time obtained from the pulse generator.

Before being used in the subsequent work, the performance of all of these new units was checked carefully, to make certain that they were functioning as was intended and were not themselves producing spectrum distortion.

Chapter 5.

The L/K Electron Capture Ratio in Chlorine-36.

Introduction:

The discussion at the end of Chapter 4 underlines the necessity for carrying out measurements of capture ratios in light isotopes. Although electron capture occurs in the decay of about 50% of the light isotopes, the choice of source is severely restricted by the need to satisfy simultaneously several conflicting conditions.

The main difficulties arise from the large differences in energy between nuclear ground states. This gives rise both to short half-lives, and to capture being in competition with beta emission. The experimental conditions are such that a minimum half-life of several hours is required and this limits the number of possible sources. Beta emission, when this is present, is usually much more intense than capture and imposes stringent demands on the apparatus, since this must be capable of discriminating against beta events while detecting capture events. Light isotopes with suitable half-lives in general decay by forbidden transitions. The systematics outlined in Chapter 1 require that the transition energy be several

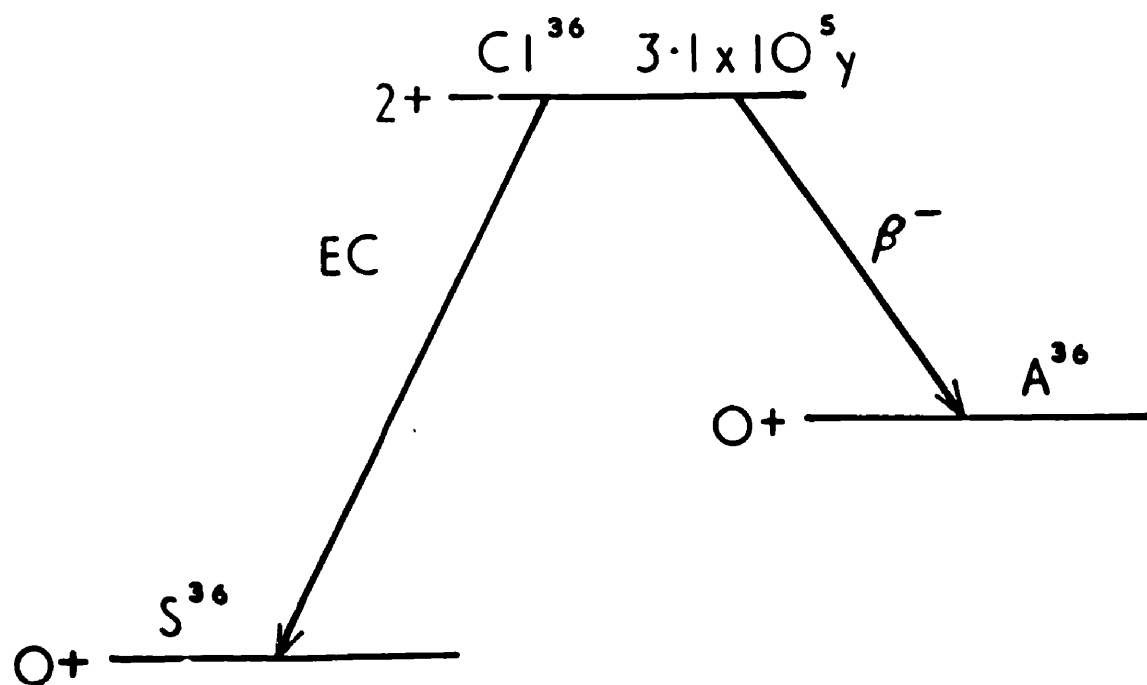


Fig.15. The decay scheme of Chlorine-36.

times the atomic K-binding energy, a requirement which is fortunately rather easy to satisfy.

Further limitations arise from the decrease of resolving power with energy which prevents any measurements being carried out on isotopes with L-binding energies much less than 200 ev, and from the necessity of obtaining the source in suitable chemical form.

As a result of a study of that part of the periodic table around mass number 40, it was concluded that the only isotopes on which electron capture ratio measurements could be carried out were A^{37} and Cl^{36} .

Results for the former were already available and have been discussed in Chapter 2. Since the technique used in these measurements was such that the escape corrections amounted to only a few per cent, and the results were in reasonable agreement with theory, there did not seem to be justification for any further measurements on A^{37} . No work on capture ratios in Cl^{36} had been reported and so attention was directed to this isotope.

The decay scheme of Cl^{36} is shown in Fig.15. The decay is of second forbidden non-unique type and so is

suitable for investigation of the form of the beta decay interaction. The spectrum shape has been studied in this connection by Wu and Feldman (1949), Fulbright and Milton (1951) and Johnson et al. (1956), the average value of the end-point energy obtained by these workers being 714 kev.

Since Cl^{36} is an odd-odd nucleus, it is expected also to be unstable against positron emission or electron capture to S^{36} . Feldman and Wu incidentally set an upper limit of 10^{-4} on positron emission relative to electron emission. The existence of an electron capture branch to S^{36} was unambiguously shown by Drever and Moljk (1956), who identified the K X-rays and Auger electrons of sulphur emitted in the atomic re-arrangement after K-capture in Cl^{36} . The ratio of K-capture to electron emission was determined to be 0.017 ± 0.001 , the capture transition being between nuclear ground states.

The energy available for electron capture in Cl^{36} has been given as 1.2 MeV by Endt and Braams (1957), so that positron emission is energetically possible. Appendix 4 contains a brief account of some scintillation

counter measurements of the spectrum of gamma-rays associated with a Chlorine-36 source. Positron emission is shown to occur with an intensity of $(11 \pm \frac{5}{2}) \times 10^{-6}$ per disintegration and the value of 1.18 ± 0.04 Mev obtained for the transition energy. At 1.2 Mev the energy available for capture is such that, according to the systematics of Brysk and Rose, the capture ratio should approximate closely to that for an allowed transition.

The L/K-ratio calculated from Brysk and Rose (1958) is 0.080 and, by analogy with the values obtained for argon, the correlations of Odier and Daudel increase this to at least 0.100.

The early measurements of Drever and Moljk (1956) used methyl chloride as source carrier and this gas was also employed in the present work.

The Cl^{36} was supplied as 2N hydrochloric acid from Amersham. Chlorine, liberated from the acid by oxidation with potassium permanganate, was mixed with methane and the mixture photolysed to form HCl and methyl chloride. Acid was removed by shaking with a dilute solution of sodium bi-carbonate, water vapour was frozen out and the methyl chloride transferred to a vessel containing a trace of mercury to remove any free chlorine liberated in the process.

Experimental Details.

The wall-less counter employed in these measurements differs from that already used. Appendix 2 contains a brief description of this counter.

Before any measurements were attempted on Cl^{36} , thorough tests of the functioning of both the counter and the associated electronics were carried out using pure electron capturing A^{37} , which emits X-rays and Auger electrons of about the same energy as Cl^{36} . These tests indicated that the counter and electronics were operating reliably: in particular, there was no difficulty in obtaining an A^{37} L-peak.

The main drawback in the experiment was the presence in the source of an electron background some 500 times stronger than the L-capture intensity. Because of the nature of the wall-less counter, the relative intensity of this background is increased by electrons passing into the central counter from the ring. The success of the counter in the Kr^{79} experiment, where there was a weak positron component, about equal to the L-capture intensity, was partly due to the high pressure, which forced the positrons to lose a large part of their total energy in the sensitive volume and so lifted these events out of

the energy region in which the capture peaks were situated, and partly to the shape of the positron spectrum which has very few low energy particles so that most of the positrons were able, even at 12 atmospheres pressure, to escape from the central counter into the ring and operate the anti-coincidence gate. In Cl^{36} , the background consists of electrons, the spectrum of which has an appreciable intensity even at low energies, and the pressure of the filling was such that the average energy loss was only a few times the K-capture energy. If the operating pressure was too high, then more of the electrons would be completely stopped in the sensitive volume and the ratio of peak to background reduced. If the pressure was too low, so that most of the electrons are removed by anti-coincidences between the counters, the escape corrections are increased. The pressure of about 1 atmosphere used endeavoured to be a compromise among these factors. A rough calculation indicated that about 95% of the total electron background would be removed by anti-coincidence and the escape corrections would be less than 10%. In fact, with all the electronic devices in use, the total background in measuring an L-spectrum was comparable to the L-peak in intensity so

LOW ENERGY SPECTRUM IN Cl^{36}

PARALYSIS TIME 400 μs

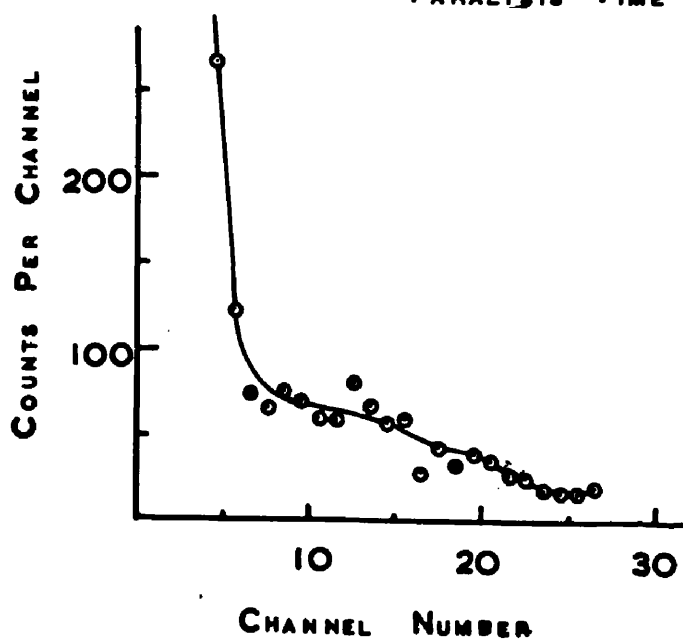


Fig.16.

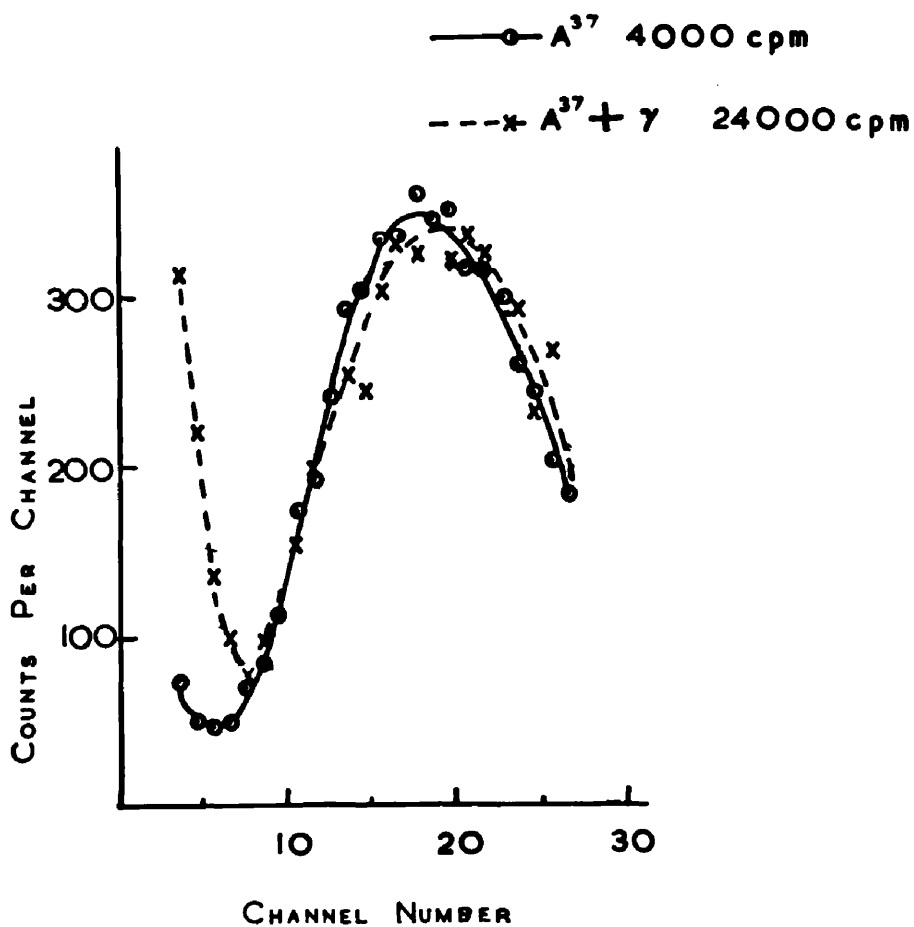


Fig.17. L-peak in A^{37} .

that the electronic circuits removed about 500 pulses for every pulse counted.

Throughout the measurements to be described, the circuits of Fig.5 were used, the gate dead-time being 150 microseconds, the delay time 20 microseconds. Initially, the paralysis unit dead-time was 400 microseconds.

The Effect of Negative Ion Formation.

On introducing labelled methyl chloride into the counter, with an argon-methane filling, it was found that though X-peaks could be obtained, the spectrum in the region where the L-peak should have occurred showed no signs of a peak, but instead rose sharply at low energies. A typical spectrum is shown in Fig.16. The peak was expected to occur about Channel 13.

Various measurements were carried out in an attempt to determine why the spectrum rose so abruptly at low energies. Eventually, the rise in the spectrum was attributed to negative ion formation. One of the initial electrons formed by an ionizing event is captured by an atom or molecule as it pulled towards the wire. Because of the low ionic drift velocities, this charge

arrives at the wire some time after the rest of the pulse has been produced and gives rise to a pulse corresponding to a single electron event. Thus the low energy spectrum measured in Cl^{36} is a combination of the beta background, the L-capture peak and the single electron spectrum arising from after-pulses. If the number of negative ions formed is comparable to the L-capture intensity, then the L-capture peak will be obscured. Since there are at least 1000 background events of perhaps 10 kev average energy for every L-event, if only 3 out of every million initial electrons form negative ions, 10 times as many after-pulses as L-events are produced.

An experimental check was carried out by a measurement in which A^{37} and an external gamma-emitting source were used to produce conditions similar to those existing in Cl^{36} .

An L-peak of A^{37} was obtained and then the gamma-source brought up towards the counter until the total counting rate had been increased by about a factor of 10. Another L-peak was taken under these conditions and both spectra normalised to approximately the same total number of counts.

These spectra are shown in Fig.17. The decrease in peak to valley ratio and the rapid rise in the number of low energy events in the presence of the gamma-source are clearly seen. These results suggest that the above interpretation of the Cl^{36} observations is correct.

Though initially introduced for other reasons, the paralysis unit of Fig.5 is clearly capable of dealing with after-pulses, since it is only necessary to insert a dead-time after every signal pulse, the length of the dead-time being longer than the time for a negative ion to drift across the counter. Though data on negative ion drift velocities is rather limited, an estimate of about 10 milli-seconds for the drift time was made for the counter dimensions and voltages used. The paralysis unit dead-time was increased to 13 milli-seconds and peaks corresponding to L-capture in Cl^{36} obtained.

With the use of the paralysis unit, it became possible to follow the shape of the energy spectrum down to the region where the counter can no longer measure energies, since it is unable to distinguish between an event caused by an electron moving only with thermal velocities and one produced with several electron volts energy.

The use of such a long dead-time is a nuisance, since the gate circuit is only open during a small part of the time. Some effort was made to eliminate the impurities responsible for negative ion formation.

First, purification of the argon-methane mixture was attempted by passing the gases over a heated copper spiral. This made no difference to the L-spectrum of Cl^{36} . Then carbon dioxide was tried as quenching gas, the gas being purified by condensation before use. Once again, no improvement was observed. In the final runs, an argon-propane mixture was used. Though this was no better than the other fillings as regards attachment, the resolution of peaks in this filling was better than with the other fillings. For instance, the resolution of a K-peak from argon-37 was 30% in an argon-methane filling and 22% in one of argon-propane. It might be remarked that because of the presence of two components in the peak, the resolution of a wall-less counter is always less good than that of a conventional counter.

The filling finally adopted was 80 cms of argon and 16 cms of propane.

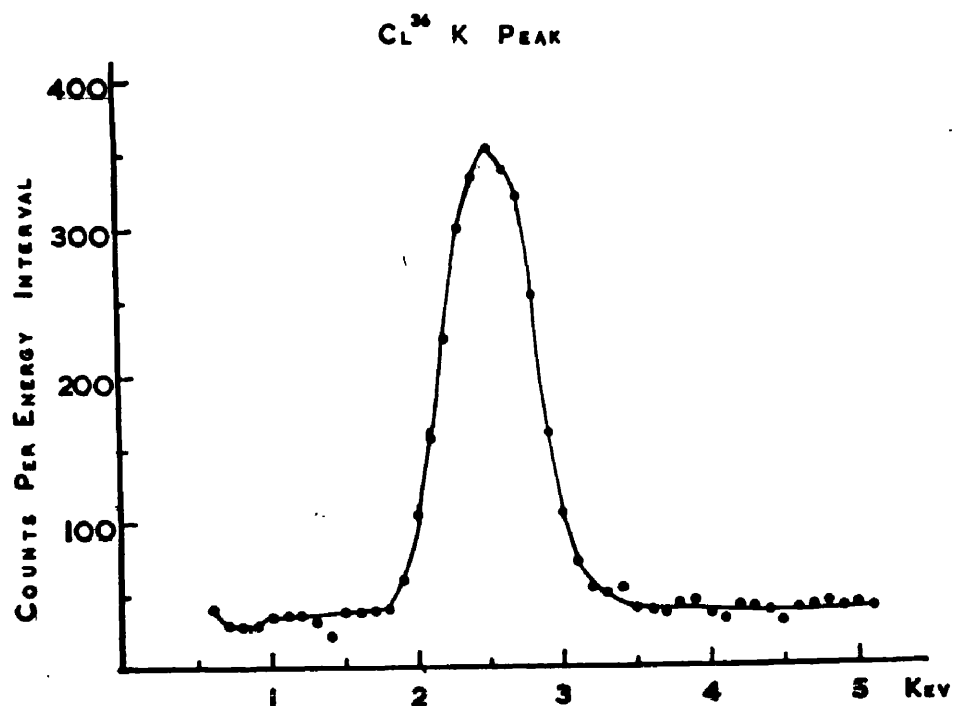


Fig.18. Cl^{36} K-peak in the central counter in anti-coincidence with events in the ring.

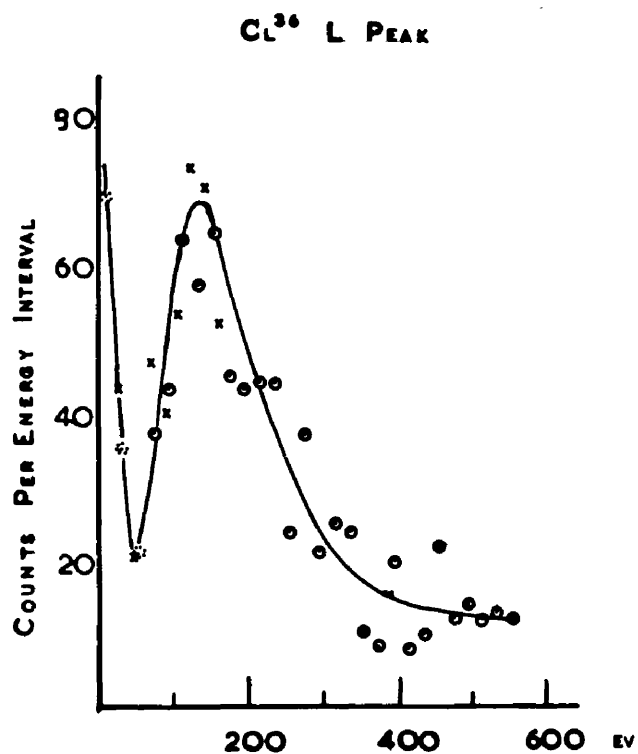


Fig.19. Cl^{36} L-peak in the central counter in anti-coincidence with events in the ring.

L/K Measurements.

To verify that 13 milli-seconds was in fact long enough to eliminate all negative ion effects, measurements of the L/K-ratio were made with various values of paralysis time between 10 and 25 milli-seconds. Agreement between the results obtained with different paralysis times was reasonable.

Figs.18 and 19 show typical K- and L-event peaks obtained with the ring bias set at 1 kev. The L-peaks comprise three separate runs taken at different amplifier gains which have been fitted together in the usual manner. Fig.20 shows a spectrum in the ring in the region of the 2.5 kev K-peak. Ten measurements of the L/K-ratio were made, the statistical error in each run being about 6%. The average value obtained was 0.117 and, after correction, the final value for the L/K electron capture ratio in Cl^{36} was 0.112 ± 0.008 . The values of $P_1 = 8 \times 10^{-3}$, $P_2 = 3 \times 10^{-2}$, $P_3 = 2 \times 10^{-2}$, $F = 0.08$ and $k = 0.9$, in the notation of (4), have been used in making the corrections.

It has been assumed that the background under an L-peak is flat and that the peak does not extend below

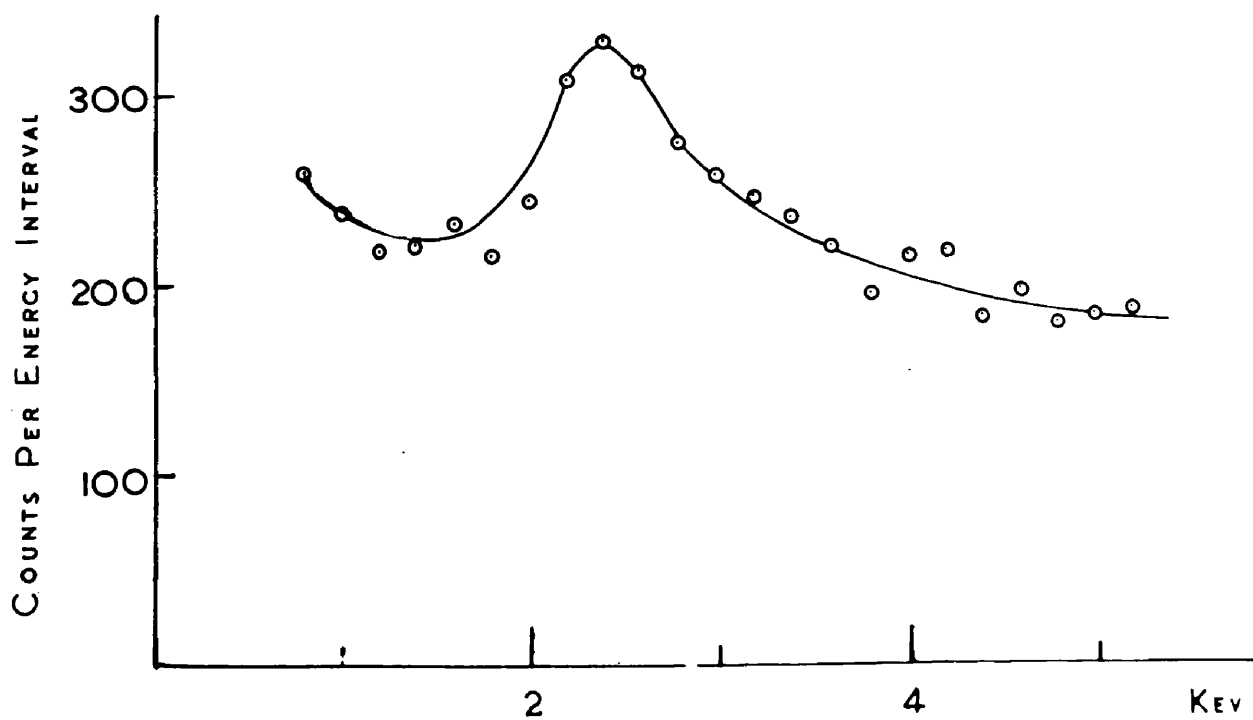


Fig.20. Spectrum of Cl^{36} events in the ring.

50 ev. With these assumptions, the ratio of peak to valley is about 6 to 1 while for the higher energy L-peak of A^{37} obtained in a separate measurement, the ratio is about 10 to 1. Agreement between peaks obtained under dissimilar conditions is thus satisfactory.

The experimental value is higher than that calculated from the work of Brysk and Rose (1958) and is comparable to the value estimated when the effect of the "correlations" is included.

This result lends support to the view that the correlation effect of Odier and Daudel is necessary. That the decay is not of allowed or unique forbidden type can hardly affect this conclusion, particularly since the density of the L_{II} and L_{III} subshells at the nucleus is very small for light elements.

Rise of the Spectrum at Low Energies.

From Fig.19, it can be seen that a rise in the spectrum of L-events was still present at low energies. This rise was found to occur even with a dead-time as long as 25 milli-seconds, and was always in the same intensity relative to the L-peak. It can hardly be explained as being due to negative ion formation. It

is tempting, in view of the recognition of M-shell capture in Ge^{71} and Kr^{79} , to attribute this rise to single electron events associated with M-capture in Cl^{36} . The question of whether a vacancy in the M_I subshell of sulphur is filled with an Auger transition or by a photo-transition is difficult to answer in view of uncertainties about the energy levels of partly excited atoms.

If the Auger effect occurs, the single electrons liberated would certainly be detected and would produce a spectrum similar in shape to that actually obtained here (Wilkinson, 1950; Curran et al., 1949). On the other hand, if photo-transitions occur, then it is not at all clear what happens to the energy, which is liberated as an ultra-violet photon. These photons may be able to ionise or may be absorbed without producing photo-electrons. In support of the view that M-capture is being observed can be quoted the fact that the intensity of the low energy part of the spectrum relative to the L-peak is about 0.14 ± 0.07 , in agreement with the value of 0.08 for the M/L ratio obtained from Fig.2.

These events could equally well arise from some other mechanism and, because of this, the above interpretation can only be considered as tentative.

It might be remarked that if M-capture is being detected, then it is possible, in principle, to detect variations in the M/L ratio with the chemical form of the source carrier, analogous to the change in decay rate observed by Leininger et al. (1951) and Louchez et al. (1949) in Be^7 and by Bainbridge et al. (1953) in $\text{Tc}^{99\text{m}}$. In practice, the effect would certainly be too small to detect.

Chapter 6.

The L/K-Electron Capture Ratio in Argon-37.

Introduction:

Argon-37 is the most suitable of all light isotopes for the study of orbital electron capture ratios. It is naturally gaseous and decays only by electron capture between nuclear ground states. The decay is allowed with $\log ft = 5.1$. The half-life is 34.5 days (Kiser and Johnston, 1960) and the transition energy has been determined as 815 kev. from nuclear reaction measurements (Richards et al., 1950), by analysis of the internal bremsstrahlung spectrum (Anderson et al., 1952; Emmerich et al., 1952; Saraf, 1956) and by the spectrometry of the charged ions produced in the capture process (Snell and Pleasanton, 1955).

The simplicity of measurements on argon-37 has produced several measurements of the L/K-capture ratio. The values are listed in Table IV.

Table IV.

Technique	L/K Capture Ratio		Reference
	Experimental	Theoretical	
Xenon filling (P = 0.13)	0.08 - 0.09	0.082 (Brysk & Rose) 0.100 (Odier & Daudel)	Pontecorvo et al. (1949)
Xenon filling (P = 0.026)	0.092 \pm 0.010 - 0.005		Langevin and Radvanyi (1955)
Argon filling (P varying from 0.4 to 1)	0.102 \pm 0.008 (F = 0.11; Haas, 1933). 0.116 \pm 0.011 (F = 0.09; Bertrand et al., 1959)		Kiser and Johnston, (1959)

In this table, P denotes the number of fluorescent K-events which escape and F is the fluorescence yield of chlorine.

The second measurement is the most accurate. Langevin and Radvanyi were able to follow the shape of the L-spectrum down to about 100 ev., at which point the ratio of peak to valley was 4 to 1. Extrapolation to lower energies was carried out by fitting a Poisson distribution to the

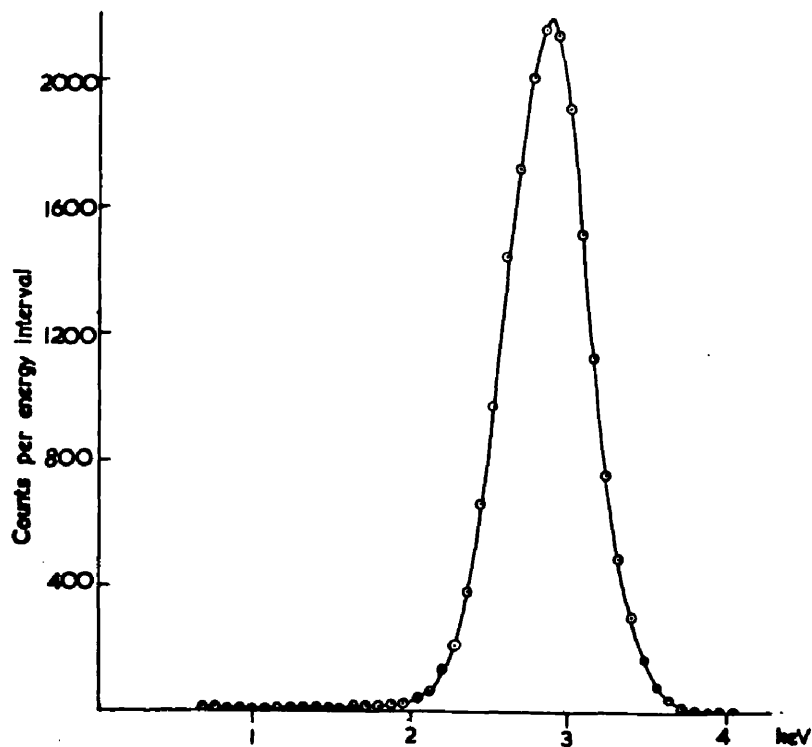


Fig.21. A^{37} K-peak in the central counter in anti-coincidence with events in the ring.

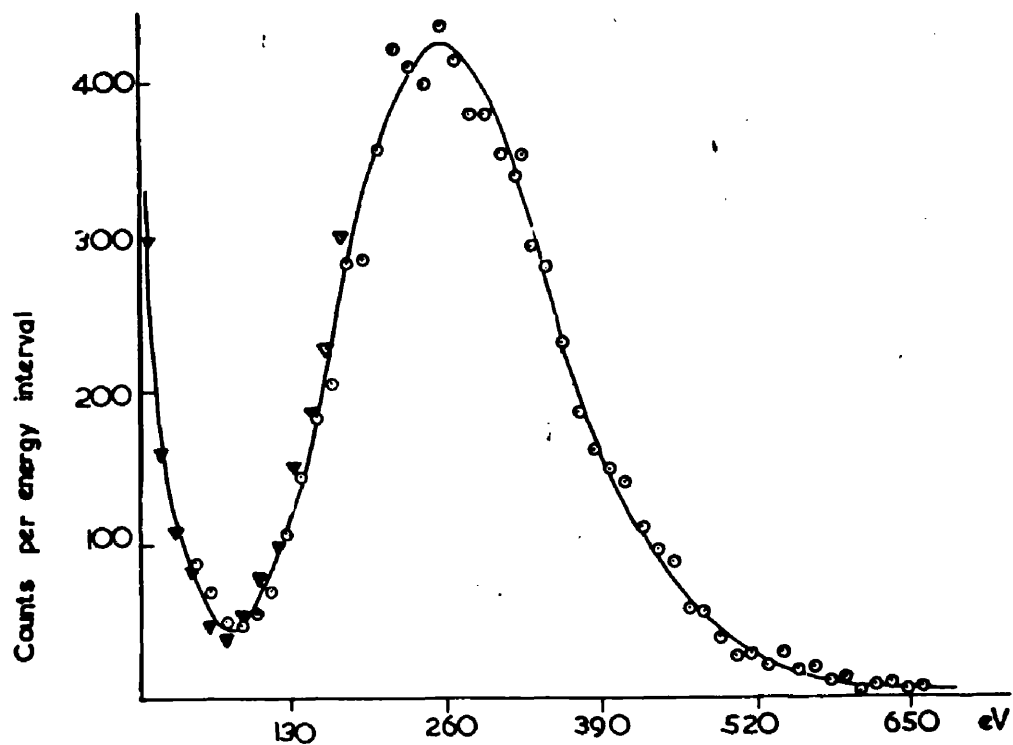


Fig.22. A^{37} L-peak in the central counter in anti-coincidence with events in the ring.

peak and is the main source of error.

The third measurement is of less importance since it involves large escape corrections and depends on the value used for the fluorescence yield.

It is seen from the above table that while these results are slightly higher than the figure computed by Brysk and Rose, the most accurate is less than the value calculated by Odier and Baudel. The techniques developed in the Cl^{36} work permit accurate measurement of spectrum shapes at much lower energies than in any of the above works and the improvement was felt to justify another measurement of the L/K-ratio in A^{37} .

Experimental Measurement.

Argon-37 source was prepared by neutron irradiation at Harwell and some of the source introduced into the multi-wire counter used in the Cl^{36} work. Experimental conditions were unchanged from that measurement.

Figs. 21 and 22 show K- and L-peaks obtained in the central counter in anti-coincidence with events greater than 1 kev. in the ring. Fig. 23 shows the spectrum in the ring in the region of the 2.8 kev K-peak.

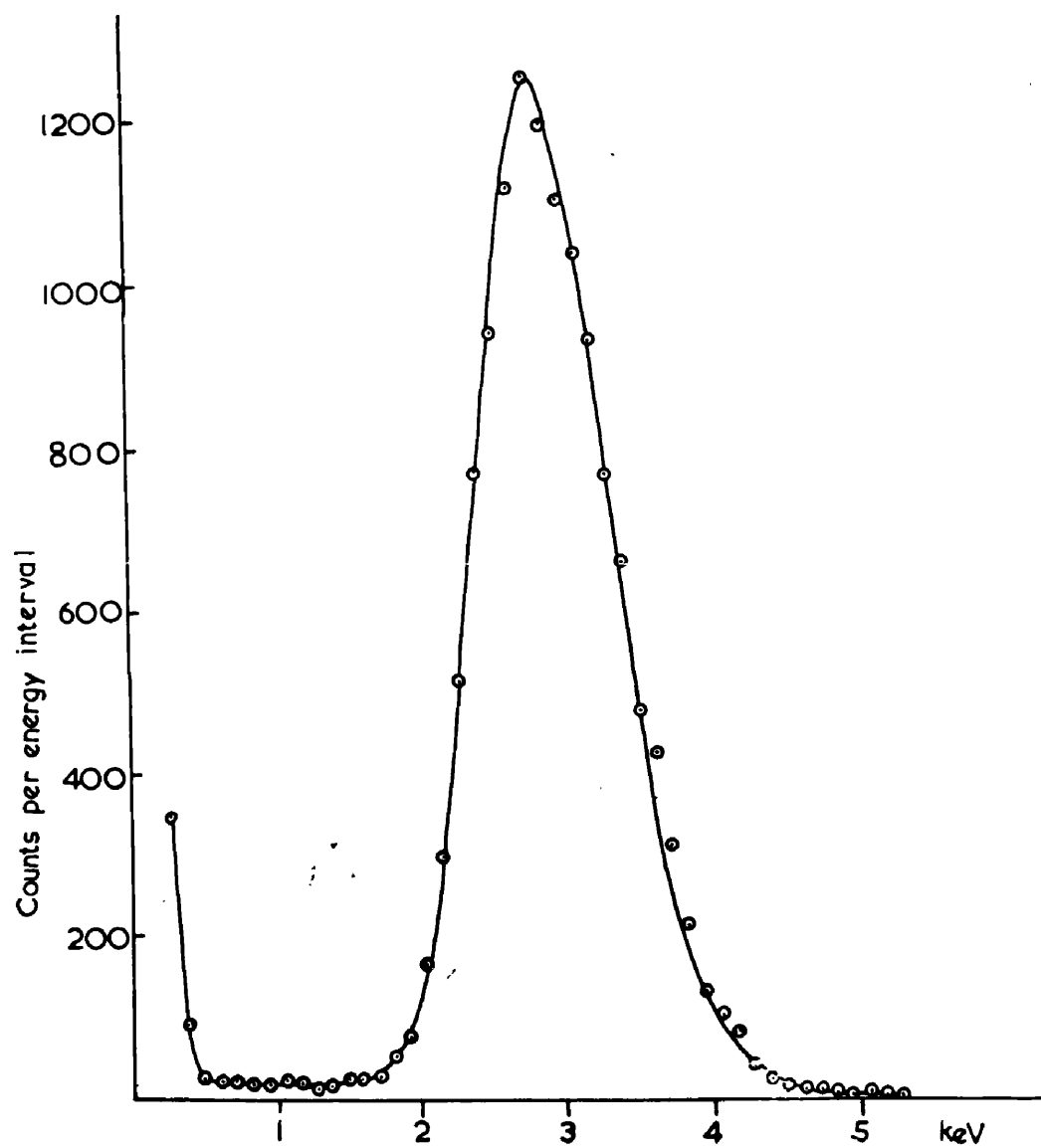


Fig.23. Spectrum of Λ^{37} K-peak in the ring.

Hardly any events occur above the K-peak in Fig.20. However, there is a slight tail on the peak and, even although the peak to valley ratio is about 250 to 1, when this tail is extrapolated linearly beneath both peaks, it contains about 2% of the total K-activity. Activity of this magnitude cannot be accounted for by any of the processes which occur with electron capture, such as internal bremsstrahlung or the "auto-ionization" of the decaying atom (Primakoff and Porter, 1953). Some of it can certainly be accounted for by the presence of impurities in the source; for instance, beta-emitting argon-41 is also prepared by neutron irradiation of argon. Natural background cannot account for more than a small fraction of the events observed, since a separate measurement made after the other runs were completed showed that such activity was less than 0.05% of the activity of the source in this energy region.

The author is of the opinion that this activity is caused by genuine K-events which are energetically degraded in some manner. It is possible that the field

tubes could be slightly non-symmetrical with respect to the anode wire, though it is worth remarking that variation of the field tube ratio over a small range about the calculated value did not have any obvious effect on the peak to valley ratio. However, on the basis of observations with the 0.003 cm. tungsten wire used for counter anodes over a wide range of conditions, the author is of the opinion that the most likely explanation for the presence of tails on peaks arises from wire non-uniformity, similar to the effect discussed in Chapter 3.

In estimating the areas of the peaks, it has been assumed that equivalent numbers of K- and L-events are degraded and the spectrum of these pulses has been assumed to be flat and to extend underneath the L-peak. Under the K-peak, the intensity has been assumed to be the average of the spectrum both above and below the peak.

The main error in the experiment arises in fixing the position of the base of the L-peak, because of the rise in the spectrum on the low energy side. Much consideration was given to ways of fitting theoretical

distributions to the upper edge of the peak to determine the true area. Explicit formulae for the shape of counter peaks do not exist and it has been customary to assume that these low energy peaks are essentially Poisson distributions. This does not seem to the author to be justified, since such distributions have only one parameter, whereas at least two parameters, corresponding to the fluctuations in ionization and in the avalanche process, are required.

The average of the 11 separate measurements of the L/K-ratio was 0.109, which, after correction for escape, gave a value of

$$0.102 \pm 0.004$$

for the L/K-capture ratio in argon-37. In the notation of equation (4), the values $P_1 = 1.3 \times 10^{-2}$, $P_2 = 4 \times 10^{-2}$, $P_3 = 3 \times 10^{-2}$, $F = 0.09$ and $k = 0.9$ have been used in correcting the observed result. Statistical accuracy in the individual runs was approximately 1%.

The result is in good agreement with the value of 0.100 calculated by Odier and Daudel. It should be remarked that this value obtained by Odier and Daudel contains only these parts of the correlations connected

with the symmetry of the wave functions, the "Pauli correlations". When all the correlations existing are included, the theoretical value is expected to be slightly higher than 0.100.

As in the previous chapter, the low energy side of the L-peak was examined with several different lengths of paralysis time. The rise in the spectrum was found to persist in the same intensity relative to the L-peak even with a dead-time of 20 milliseconds.

The measurements carried out under the highly dissimilar conditions of the work on Cl^{36} and A^{37} both show this rise in the low energy spectrum and, in each case, the intensity of the rise relative to the L-peak is constant over a wide range of paralysis time. The careful tests of reliability of the electronic units remove all possibility of this rise being associated with electronic faults, a deduction which is confirmed by the manner in which spectra taken at different amplifier gains fit together.

The only possible causes of this rise are either a common origin of single electron events in both sources or else some mechanism in the counter which also produces

single electrons. There is no obvious mechanism of this type in the counter, and hence it seems reasonable to conclude that the common origin of the rise is the true explanation. M-capture is then the simplest explanation.

As in the case of Cl^{36} , there are uncertainties as to whether or not the Auger effect is possible, subsequent to M-capture. Snell and Pleasonton (1955) assume that M Auger events occur, to account for the existence of recoil ions of charge +7 units, but they point out that because of the virtually complete lack of knowledge about the energy levels of excited atoms and ions, it is not known whether such transitions are energetically possible. If energetically possible, then because of the considerable overlap of the relevant wave functions, the Auger transition would occur with a high probability (Burhop, 1952).

The intensity of the rise relative to the L-peak, after correction for the escape of K X-rays is 0.08 ± 0.04 , in good agreement with the value of 0.08 for the M/L-capture ratio obtained from Fig.2.

By calibrating the relative amplifier gains with a pulse generator, it was possible to make an estimate of the mean energy of the L-peak. This was found to be 275 ± 10 ev, considerably higher than the value of 238 ev obtained from X-ray absorption edges by Hill et al. (1952). However, recently a value of 268 ± 8 ev has been given for the energy release subsequent to L-capture in A^{37} (Santos-Ocampo and Conway 1960), which is in better agreement with the above result. It must be pointed out that the above measurement is not precise, since, although the energy of the K-peak has been taken as 2.82 kev, it is in fact slightly lower than this value because of the presence of two components in the K-spectrum. In addition to this, the observed L-peak contains a few per cent of L_{II} or L_{III} events, arising from the escape of K_{α} X-rays. However, the mean energy release after an L-capture transition in argon-37 should lie within the range of values quoted above.

Some time after these results had been obtained, the result of a measurement of the L/K-ratio in A^{37} was

published by Santos-Ocampo and Conway (1960), their value of 0.103 ± 0.003 being in excellent agreement with the above value. These workers used a multi-wire counter technique similar to the above, although, since they were primarily interested in measuring the average energy of the L-peak, they used much higher pressures to minimise the ambiguity in pulse height inherent in the technique. Though these workers do not explicitly mention the presence of afterpulses, they were unable to obtain L-peaks without thorough purification of the gases prior to filling the counter.

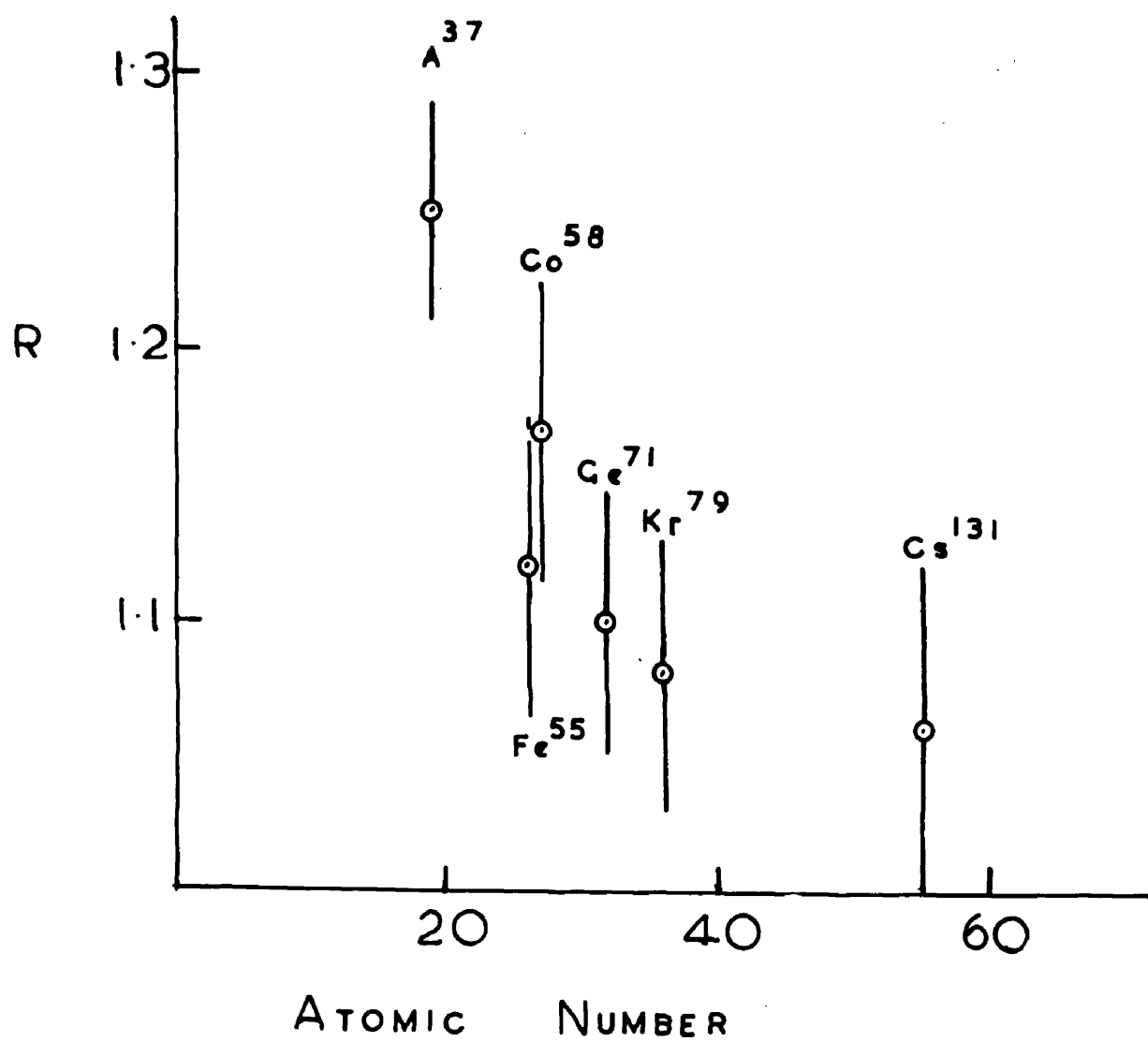


Fig.24.

Chapter 7.

Discussion.

In the period since the compilation of Table II, several new values of L/K-ratios, suitable for comparison with theory, have been published. Those values reported up to April 1961, together with those obtained in the present work, are listed in Table V. (See overleaf).

In Fig.24, R, the ratio of the experimental value of the L/K-ratio to the theoretical value, has been plotted as a function of atomic number.

At high values of atomic number, the only experimental point, that of thallium-204, is in good agreement with theory, although the energy dependence of the L/K-ratio in this isotope is such that a more accurate measurement of the transition energy would be of value.

The excellent agreement between the value calculated by Odier and Daudel for argon-37 and the experimental value, taken in conjunction with the general trend in experimental values revealed in Fig.24, strongly suggests that the effects of electron correlation considered by

Table V.

Isotope	L/K-Ratio		M/L-Ratio		Technique	Comment	Reference
	Experimental	Theoretical	Experimental	Theoretical			
Cl^{36}	0.112 ± 0.008	0.080			Wall-less counter	Second-forbidden non-unique decay	Chapter 5
La^{37}	0.103 ± 0.003	0.082 (Brysk and Rose)			do.	Allowed transition	Santos-Ocampo and Conway (1960)
	0.102 ± 0.004	(Odier and Daudel)			do.		Chapter 6
Fe^{55}	0.106 ± 0.003	0.095			do.	First-forbidden transition	Scobie, Moler & Fink (1959).
							Moler & Fink (1961)
Co^{58}	0.108 ± 0.004	0.092			do.	Allowed transition	Moler and Fink (1961)
Ge^{71}	0.116 ± 0.005	0.106	0.16 ± 0.08	0.153	do.	Allowed transition	Chapter 3
Kr^{79}	0.108 ± 0.005	0.101	0.16 ± 0.08	0.16	do.	Allowed transition	Chapter 4
Cs^{131}	0.153 ± 0.008	0.145			Scintillation counter	Allowed transition	Joshi and Lewis (1960)
Tl^{204}	0.42 ± 0.05	0.45 ± 0.04			do.	Transition energy taken as 380 - 20 kev. (Jung & Pool, 1956)	Joshi (1961)
Np^{235}			0.35 ± 0.04	0.32	Proportional counter		Gindler et al. (1958)

these authors need to be examined in more detail; in particular, some effort should be made to determine the relative magnitude of the correlations as a function of atomic number, over a wider range of atomic number than was considered by Odliot and Daudel.

It is possible that, when such a determination has been made, the small differences now remaining between the theoretical and experimental values for isotopes such as iron-55 and krypton-79 could be accounted for.

Also included in Table V are the three available values of M/L-ratios, which are given here as electron density ratios, derived by removing the energy dependent factor.

Agreement between the experimental results and the theoretical values derived from Fig.2 is seen to be reasonable, within the limited accuracy of the experiments.

It should be remarked that the technique developed to remove after-pulse effects in the work on chlorine-36 could be used to obtain more accurate values of the M/L-ratios for germanium-71, krypton-79 and other isotopes in this region of the periodic table, while the whole technique could be employed to measure M/L-ratios at the

upper end of the periodic table in those isotopes, such as lead-202, in which decay by K-electron capture is energetically forbidden.

In conclusion, it may be said that the combination of the wall-less counter technique with the electronic circuits of Fig.5 has been shown in the present thesis to be of unrivalled power and versatility for the measurement of orbital electron capture ratios in isotopes with mass numbers less than 100.

Appendix 1.

Data Pertaining to Wave Functions.

(a) Marshak's Wave Functions.

The analytical forms of the wave functions for an electron in the Coulomb field of a point nucleus given by Marshak (1942) are:

$$g_K^2 = \frac{1 + W_K}{2 \Gamma(2s_0 + 1)} (2\alpha Z_{\text{eff}} R)^3 \exp(-2\alpha Z_{\text{eff}} R) (2\alpha Z_{\text{eff}} R)^{2s_0-2}$$

$$g_{L_I}^2 = \frac{(2s_0 + 1)(2 + 2s_0)^{1/2} (1 + W_{L_I})}{4 \Gamma(2s_0 + 1) [(2 + 2s_0)^{1/2} + 1]} \left(\frac{2\alpha Z_{\text{eff}} R}{(2 + 2s_0)^{1/2}} \right)^3$$

$$\exp\left\{-\frac{2\alpha Z_{\text{eff}} R}{(2 + 2s_0)^{1/2}}\right\} \cdot \left(\frac{2\alpha Z_{\text{eff}} R}{(2 + 2s_0)^{1/2}}\right)^{2s_0-2}$$

with $s_0 = \left(1 - \alpha^2 Z_{\text{eff}}^2\right)^{1/2},$

$$\alpha = \frac{1}{137},$$

$$R = 1.2 A^{1/3} \times 10^{-13} \text{ cm.}$$

and, for K-electrons, $Z_{\text{eff}} = Z - 0.3$

L-electrons, $Z_{\text{eff}} = Z - 4.15.$

Marshak gives the values of W_{LI} and W_K , the binding energies of a bound electron, as

$$W_K = \left(1 - \frac{\alpha^2 Z_{eff}^2}{2}\right), \quad W_{LI} = \left(1 - \frac{\alpha^2 Z_{eff}^2}{8}\right)$$

but in preparing values, the experimental binding energies have been used in preference to these approximate values.

(b) Hartree Wave Functions.

The Hartree wave functions used in preparing Figs. 1 and 2 are contained in the following references:

- 1) N, Na: Hartree, D.R. and Hartree W., 1948, Proc. Roy. Soc. A193, 299.
- 2) A : Hartree, D.R. and Hartree, W., 1938, Proc. Roy. Soc. A166, 450.
- 3) Zn : Hartree, D.R., Hartree, W. and Manning, M.F., 1941, Phys. Rev. 59, 299.
- 4) Mo : Ridley, E.C., 1955, Proc. Cambridge Phil. Soc. 51, 702.
- 5) Cs, K: Hartree, D.R., 1933, Proc. Roy. Soc. A143, 506.
- 6) Tl, Au: Douglas, A.S., Hartree, D.R. and Runoiman, W.A., 1955, Proc. Cambridge Phil. Soc. 57, 486.

General reviews of the calculation of Hartree wave functions have been given by Hartree (1946, 1957).

Appendix 2.

Details of the Counters Used in the Measurements.

1. The counter used for measurements with Ge⁷¹ and Kr⁷⁹.

This counter, of total volume 20 litres, was designed to withstand operating pressures up to 15 atmospheres. It is shown diagrammatically in Fig.3.

The central counter, 4.5 cm. in diameter and 76 cm. in sensitive length, is fitted with both end-correcting and guard tubes.

Five separate counters, defined by 15 0.020 cm. stainless steel wires, form the ring system. The anode wires of these counters are protected by guard tubes and are connected together internally, an annular box providing screening. The whole ring assembly is mounted between brass plates separated by five brass rods and can be removed from the 18 cm. diameter brass case as a unit, so that the fitting of the wires is relatively simple.

The six tungsten anode wires are all 0.003 cm. in diameter.

At 12 atmospheres pressure, the operating voltage is about 7 kv.

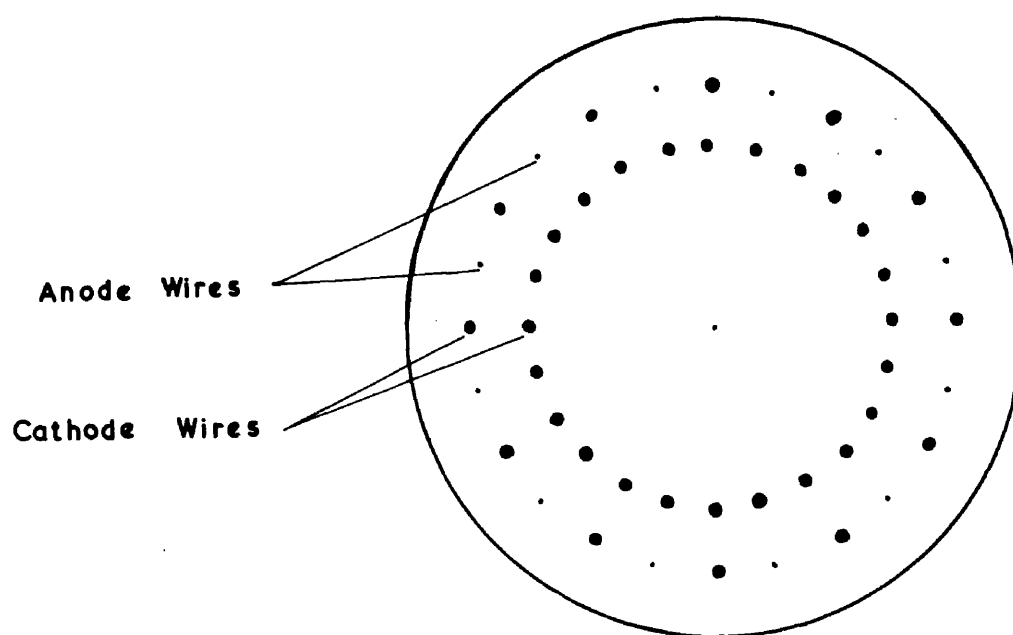
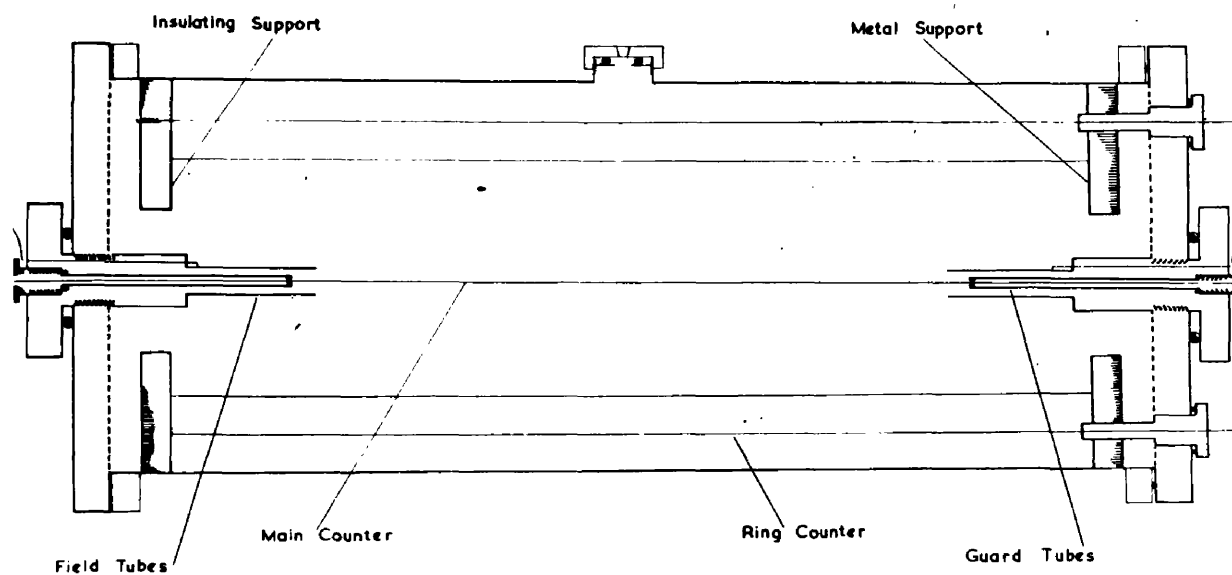


Fig.25. The wall-less counter used in measurements on Cl^{36} and A^{37} .

2. The counter used for measurements on Cl^{36} and A^{37} .

This counter is a modification of a counter initially designed by Scobie (1957). It is shown diagrammatically in Fig.25.

The ring system, 1.6 cm. thick, contains 36 cathode wires, 0.013 cm. in diameter, connected to the case. The 12 anode wires, 0.005 cm. in diameter, are brought out through perspex insulators and connected together externally. The ring system has neither field tubes nor guard tubes. Despite this, no difficulty was experienced from sparking.

The central counter has a mean diameter of 4.5 cm. and a sensitive length of 45 cm. It is fitted with both guard tubes and field tubes. The anode wire is 0.003 cm. in diameter and, like all the other wires in this counter, is of tungsten.

At 1 atmosphere, the operating voltage was about 2 kv., and, with the above choice of wire diameters, the pulses from both the ring and the central counters were approximately equal in amplitude, so that no provision for separate bias of the ring was necessary.

Appendix 3.

Some Remarks on the Electronic Units.

The greater part of the gate circuit, which is shown in the form of a block diagram in Figs. 4 and 5, and as a circuit diagram in Fig.26, consists of standard circuits, and so does not require description. However, it should be remarked that the gate circuit was designed so that paralysis effects arising from overload are minimised. For this reason, the circuits are D.C. coupled wherever possible, and the pulses from the counter are doubly-differentiated, rather than the more usual singly-differentiated, since this practice greatly reduces the degree of overload arising from a very large pulse.

A circuit diagram of the new units added is shown in Fig.27. Valves V22, V30, V23a and V24 to V26 are the paralysis unit and valves V4b, V23b and V28 form the coincidence unit.

The detailed operation of the paralysis unit is most clearly seen by showing the waveforms at various points, and this has been carried out in Fig.28, in

which it is seen how a pulse is transmitted before the paralysis time is applied.

The coincidence unit is a standard diode coincidence unit. If pulses are present on both the veto and the signal sides of the gating valve, a coincidence signal produced, and since this is fed back to the mixer, the gate is prevented from opening.

Appendix 4.

Positron Emission in the Decay of Chlorine-36.

The transition energy for electron capture in chlorine-36 has been calculated to be 1.2 MeV from atomic mass differences by Endt and Braams (1957). This value is greater than the threshold for positron emission and, during the measurements of the L/K-ratio, it was decided to make a brief study to see if a positron decay branch could be detected.

Previous investigations of positron activity had been carried out by Feldman and Wu (1949), who set an upper limit of $\beta^+/\beta^- < 10^{-4}$ for positron emission relative to electron emission, and by Johnson and Willard (1949), who claimed to have detected positron emission of intensity $\beta^+/\beta^- = 3 \times 10^{-4}$. Considerable difficulties of interpretation occur with the type of differential gamma-ray absorption technique used by these latter authors, and it is probable that their results are not too reliable.

Since the simplest way of recognising positrons is by detection of the 0.511 MeV quanta produced on

APPARATUS FOR β^+ IN Cl^{36}

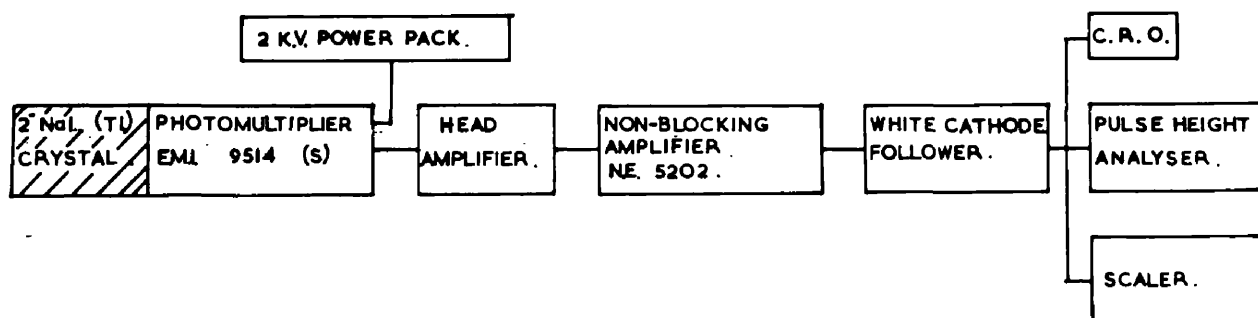


Fig.29.

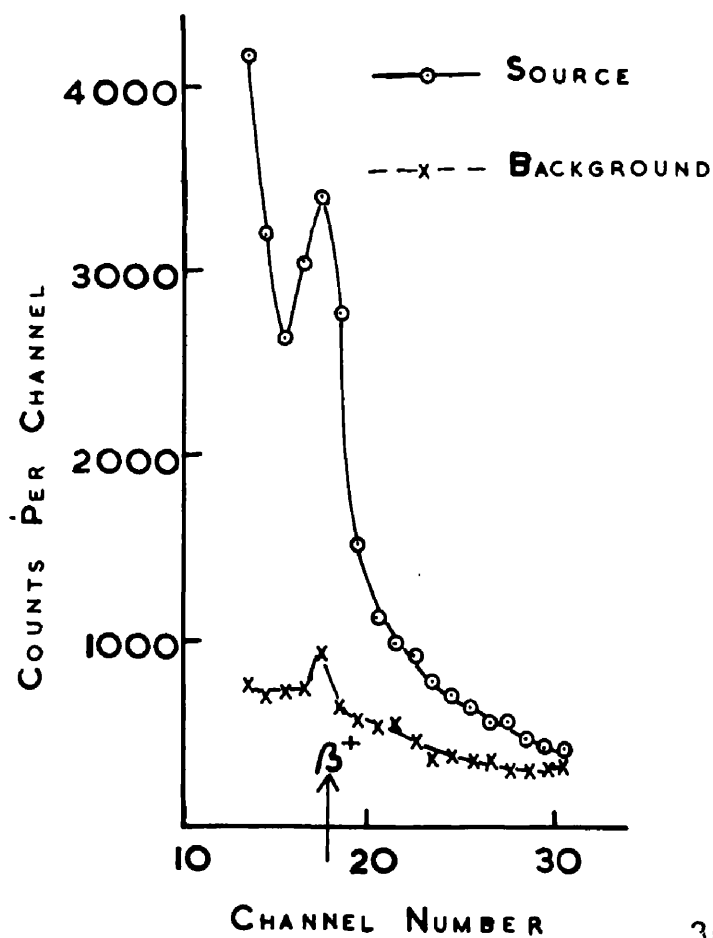


Fig.30. The gamma-ray spectrum of Cl^{36} between 0.40-1 Mev.

annihilation, the investigation was carried out by examining the gamma-ray spectrum emitted by a 15 micro-curie source of chlorine-36, supplied as 2N HCl from Amersham.

A 5 cm. by 5 cm. NaI(Tl) crystal was mounted on an EMI 9514 (S) photo-multiplier, and, to reduce background, enclosed in a lead castle of minimum wall thickness 4 inches. The output pulses were amplified on an NE 5202 (A) non-overloading amplifier and analysed by a Marshall kicksorter. The layout is shown in Fig.29. Measurements with the source present were alternated with measurements of the background. Typical spectra in the 0.5 MeV energy region are shown in Fig.30. Both with and without the source present, there is a peak at about 0.5 MeV. D'Angelo (1959) has suggested that the peak in the background is due to excitation of a level at 0.5 Mev in the lead castle by the fast neutron component of the cosmic rays. The peak obtained with the source present is some 12 times greater in intensity. It is superimposed on a rapidly rising background caused by external bremsstrahlung from the electron component of the source. Energy calibration and the estimation of the relative areas under the peaks were carried out by

β^+ COINCIDENCE ARRANGEMENT IN ^{36}Cl

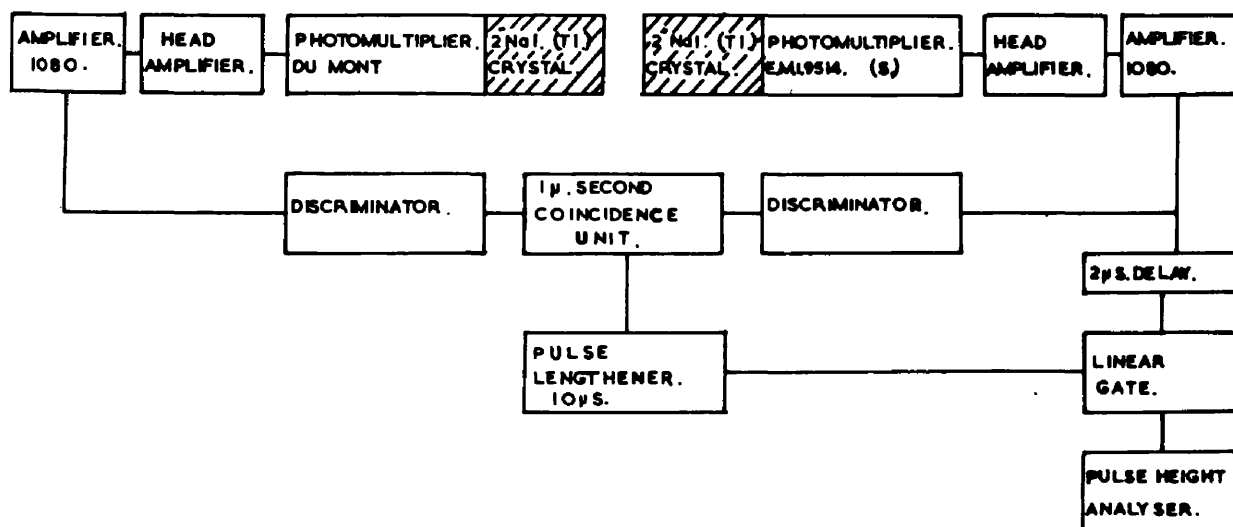


Fig.31.

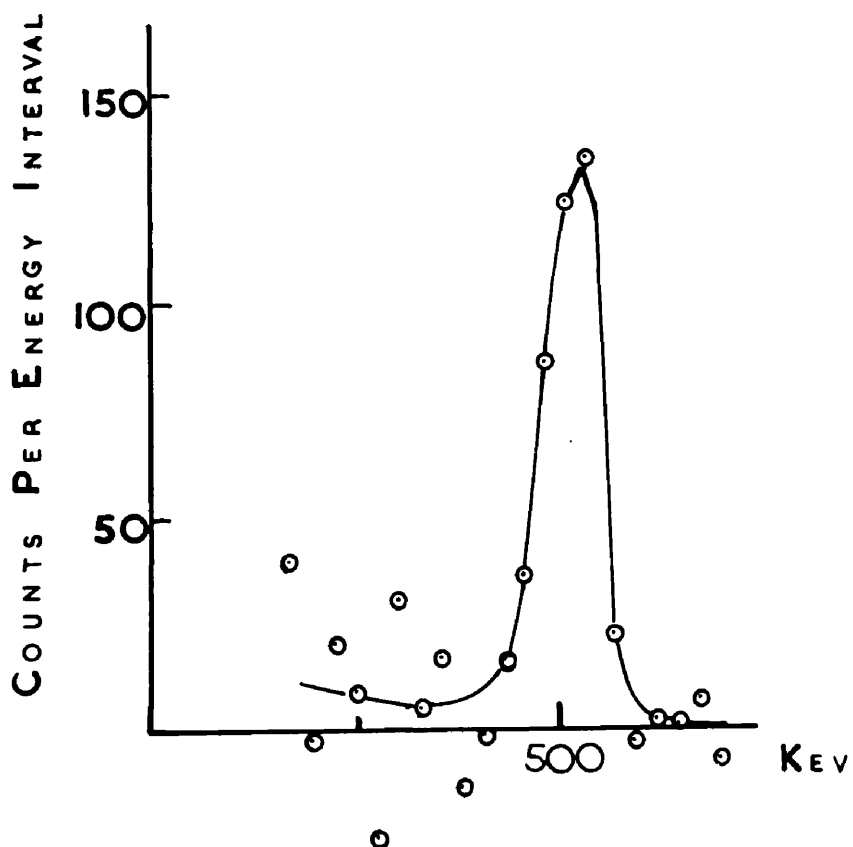


Fig.32. The coincidence spectrum.

comparison with the positron annihilation quanta from a sodium-22 source.

The peak obtained with the source present can be explained in several ways. It can arise from a weak branching to a hitherto unreported level at 0.5 Mev in argon-36 or sulphur-36, it can be due to the presence of a small amount of a positron-emitting impurity in the source, or it could be associated with the chlorine-36.

Since the annihilation of a positron produces two 0.5 Mev quanta in opposite directions to each other, the existence of a level at 0.5 Mev can be tested by an angular correlation study. The layout used is shown in Fig.31, and is a standard coincidence unit. Three measurements were carried out. In the first two, the coincidence spectrum with the detectors 180' apart, on either side of the source, was measured. The discriminator levels in both these runs were set at 100 kev on the signal channel and at 400 and 600 kev on the other channel in the first and second run respectively. In the third run, one of the detectors was moved through 90'. The levels were again 100 and 400 kev. The first two runs define an energy range in which the coincidence

- 87 -

event must lie, while the third gives a measurement of the spectrum due to randoms.

Fig.32 shows the difference between the first and third runs. The peak observed agreed in energy and shape with a peak given by sodium-22 under similar circumstances.

It is reasonable to conclude that these measurements confirm the existence of a positron emitter in the source.

Some indication that the observed positron activity was due to the chlorine-36 and not to the presence of an impurity was given by measurements of the gamma-ray spectrum emitted by a chlorine-36 source which had been supplied from Chalk River in 1956. This source gave spectra similar to that in Fig.30, both in the position of the peak and in its intensity. Further indication of the purity of the sources was derived by examining the spectra for the presence of other gamma-rays. None were found.

These observations do not provide unambiguous confirmation that the positrons arise from chlorine-36. This was emphasised when a 60 micro-curie source was obtained from Amersham. It was found to contain some

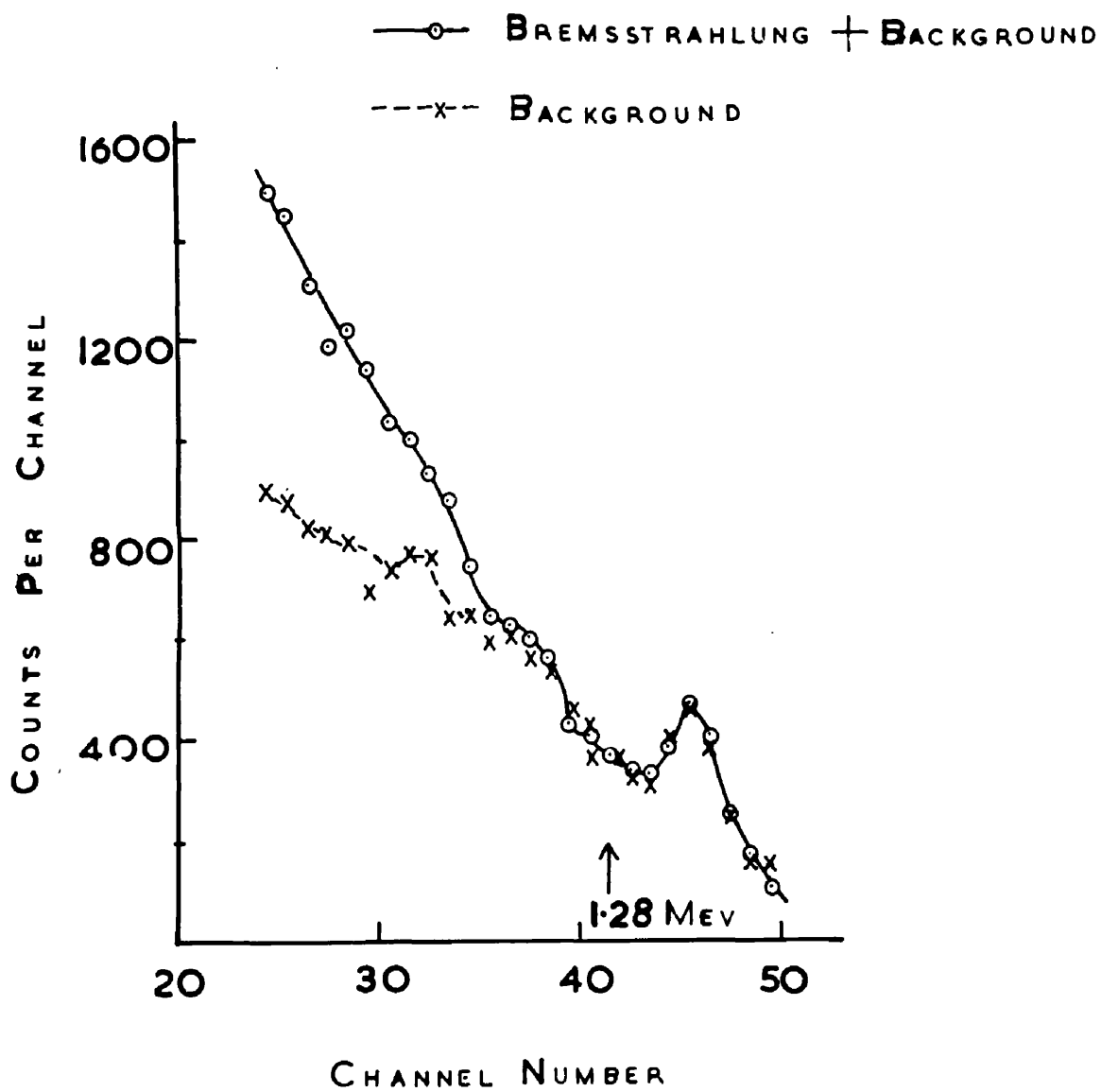


Fig.33. The gamma-ray spectrum of Cl^{36} above 0.7 Mev.

impurity which emitted two gamma-rays of about 1.2 Mev, in addition to positrons in about the same intensity as the other two sources.

The most satisfactory manner of obtaining unequivocal confirmation that the observed activity is associated with the chlorine-36 is to carry out a chemical separation and purification of the chlorine. The weakness of the positron events relative to the external bremsstrahlung makes such a procedure very difficult here, since, to permit an accurate comparison of the intensity of the peak both before and after purification, it would be necessary for the initial and final form of the source to be identical. (The external bremsstrahlung is a function of the atomic number of the surrounding medium.)

Further evidence that the positrons originate in the chlorine was obtained by a measurement of the internal bremsstrahlung associated with electron capture (Wu, 1955).

The same experimental arrangement as in Fig.29 was used; Fig.33 shows the gamma-ray spectrum of the source and the background in the 1 Mev energy region. The spectrum with the source present is seen to converge to that of the background in the 1.2 Mev energy region. Though the intensity of the external bremsstrahlung

prevented measurements of the spectrum shape over a wide range of energy, it was possible to conclude that the transition energy for electron capture in chlorine-36 is

$$1.18 \pm 0.04 \text{ Mev.}$$

The peak at 1.45 Mev in Fig.33 is due to potassium-40 contamination in the NaI(Tl) crystal.

While no single one of the above observations conclusively proves that the positron activity observed originates in chlorine-36, the consistency of the results appears to favour this interpretation. The value obtained for the transition energy indicates that positron emission is energetically possible, and the intensity of positron emission of $(11 \pm 2) \times 10^{-6}$ per disintegration seems not unreasonable for a second-order forbidden transition of 160 kev end-point energy. Accurate comparison of the theoretical and experimental values of the K/β^+ ratio of 1500 ± 300 cannot be made without knowledge of the nuclear matrix elements involved.

In calculating the above intensity, the source has been assumed to be a point source on the axis of the

crystal, though it was in the form of a right circular cylinder. The values of 0.10 and 0.4 have been used for the total detection efficiency and the photo-peak efficiency of a 5 cm. by 5 cm. NaI(Tl) crystal with an axial source at an average distance of 1.5 cm. (Mott and Sutton, 1958).

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Summary.

**Proportional Counter Studies
of
Orbital Electron Capture Ratios**

by

P.W. Dougan.

Ph.D. Thesis: May, 1961.

Summary.

The thesis commences with a brief review of the theory of electron capture. A discussion is given of the atomic electron wave functions available for the calculation of electron capture ratios and it is shown that agreement between the few experimental measurements of L/K-capture ratios and the theoretical values computed by Brysk and Rose is poor. The necessity for further experimental work is emphasised.

The second chapter is devoted to an account of the methods available for measurements on isotopes with mass number less than 100. The wall-less counter method developed by Drever and Moljk is shown to be a major advance in technique.

The next four chapters illustrate the application of wall-less counter methods to the measurement of L/K-ratios in the isotopes germanium-71, krypton-79, chlorine-36 and argon-37. The experimental values of 0.116 ± 0.005 , 0.108 ± 0.005 , 0.112 ± 0.008 and 0.102 ± 0.004 are all slightly larger than the respective theoretical values of 0.106, 0.101, 0.081 and 0.082 calculated from the data of Brysk and Rose. The experimental value for argon-37 is in good agreement with a theoretical value of 0.100 calculated by Odier and Daudel in which consideration is

given to the effect of correlations existing among the atomic electrons.

The occurrence of electron capture from the M-shell is shown unambiguously for germanium-71 and krypton-79, the first direct observation of M-capture in isotopes with mass number less than 200. The value of 0.16 ± 0.08 for the M/L ratio in both isotopes is in agreement with values of 0.15 and 0.16 calculated from Hartree wave functions.

The presence of M-capture in chlorine-36 and argon-37 is inferred from observations made in the measurements on these isotopes, but the evidence obtained is not conclusive

The final chapter summarizes ^{various} ~~the~~ points of counter technique clarified by the measurements and contains a discussion of the present status of the theoretical calculations of electron capture ratios in the light of the above and other recent measurements. It is concluded that further theoretical work to evaluate the effect of electron correlations is required.

An appendix deals with scintillation counter measurements of the gamma-ray spectrum associated with electron

capture in chlorine-36. A weak positron branching of intensity $(7 \pm 3) \times 10^{-4}$ relative to K-capture is detected and the energy available for the electron capture transition is found to be 1.18 ± 0.04 MeV.